Superconductivity

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Perovskite and Cuprate Crystallographic Structures

I. INTRODUCTION

Chapter 3 shows that the majority of single-element crystals have highly symmetrical structures, generally fcc or bcc, in which their physical properties are the same along the three crystallographic directions x, y, and z. The NaCl-type and $A15$ compounds are also cubic. Some compounds do have lower symmetries, showing that superconductivity is compatible with many different types of crystallographic structure, but higher symmetries are certainly more common. In this chapter we will describe the structures of the high-temperature superconductors, almost all of which are tetragonal or orthorhombic.

In Chapter 3, we also gave some examples of the role played by structure in determining the properties of superconductors. The highest transition temperatures in alloys of transition metals are at the boundaries of instability between the bcc and hcp forms. The NaCl-type compounds have ordered vacancies on one or another lattice site. The magnetic and superconducting properties of the Chevrel phases depend on whether the large magnetic cations (i.e., positive ions) occupy eightfold sites surrounded by chalcogenide ions or whether the small magnetic ions occupy octahedral sites surrounded by Mo ions.

The structures described here are held together by electrons that form ionic or covalent bonds between the atoms. No account is taken of the conduction electrons, which are delocalized over the copper oxide planes and form Cooper pairs responsible for the superconducting properties below T_c. The following chapter will be devoted to explaining the role of these

conduction electrons within the frameworks of the Hubbard model and band theory. Whereas the present chapter describes atom positions in coordinate space, the following chapter relies on a reciprocal lattice elucidation of these same materials.

We begin with a description of perovskite and explain some reasons that perovskite undergoes various types of distortions. This prototype exhibits a number of characteristics that are common to the high-temperature superconducting cuprates (see Section V). We will emphasize the structural commonalities of these materials and make frequent comparisons between them. Our earlier work (Poole *et al.*, 1988) and the comprehensive review by Yvon and François (1989) may be consulted for more structural detail on the atom positions, interatomic spacings, site symmetries, etc., of these compounds. There have been reports of superconductivity in certain other cuprate structures (e.g., Murphy *et al.*, 1987), but these will not be reported on in this chapter.

There is a related series of layered compounds $Bi_2O_2(M_{m-1}R_mO_{3m+1})$ called Aurivillius (1950, 1951, 1952) phases, with the 12-coordinated M = Ca, Sr, Ba, Bi, Pb, Cd, La, Sm, Sc, etc., and the 6-coordinated transition metal R = Nb, Ti, Ta, W, Fe, etc. The $m = 1$ compound Bi_2NbO_6 belongs to the same tetragonal space group $I4/mmm$, D_{4h}^{17} as the lanthanum, bismuth, and thallium high temperature superconductors (Medvedeva *et al.*, 1993).

We assume that all samples are well made and safely stored. Humidity can affect composition, and Garland (1988) found that storage of $YBa_2Cu_3O_{7-\delta}$ in 98% humidity exponentially decreased the diamagnetic susceptibility with a time constant of 22 days.

II. PEROVSKITES

Much has been written about the high-temperature superconductors being perovskite types, so we will begin by describing the structure of perovskites. The prototype compound barium titanate, $BaTiO_3$, exists in three crystallographic forms with the following lattice constants and unit cell volumes (Wyckoff, 1964):

cubic:	$a = b = c = 4.0118$ Å	$V = 64.57$ Å^3	
tetragonal:	$a = b = 3.9947$, $c = 4.0336$	$V = 64.37$ Å^3	(7.1)
*ortho*rhombic:	$a = 4.009\sqrt{2}$ Å, $b = 4.018\sqrt{2}$ Å, $c = 3.990$ Å	$V = 2(64.26)$ Å^3	

For all three cases the crystallographic axes are mutually perpendicular. We will comment on each case in turn.

A. Cubic Form

Above 201°C barium titanate is cubic and the unit cell contains one formula unit $BaTiO_3$ with a titanium atom on each apex, a barium atom in the body center, and an oxygen atom on the center of each edge of the cube, as illustrated in Fig. 7.1. This corresponds to the barium atom, titanium atom, and three oxygen atoms being placed in positions with the following x, y, and z coordinates:

E site: Ti	(0,0,0)	Ti on apex	
F site: O	$(0,0,\frac{1}{2})$; $(0,\frac{1}{2},0)$; $(\frac{1}{2},0,0)$	three oxygens centered on edges	(7.2)
C site: Ba	$(\frac{1}{2},\frac{1}{2},\frac{1}{2})$	Ba in center.	

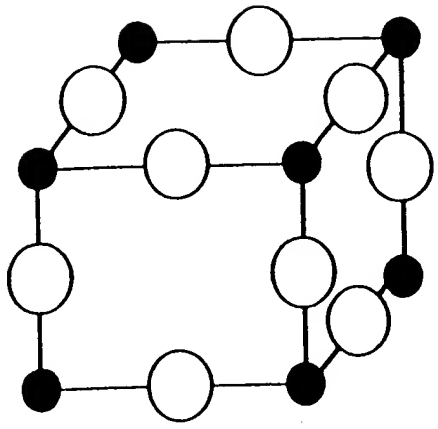


Figure 7.1 Barium titanate ($BaTiO_3$) perovskite cubic unit cell showing titanium (small black circles) at the vertices and oxygen (large white circles) at the edge-centered positions. Ba, not shown, is at the body center position (Poole *et al.*, 1988, p. 73).

The barium in the center has 12 nearest-neighbor oxygens, so we say that it is 12-fold coordinated, while the titanium on each apex has 6-fold (octahedral) coordination with the oxygens, as may be seen from the figure. (The notation E for edge, F for face, and C for center is adopted for reasons that will become clear in the discussion which follows.) Throughout this chapter we will assume that the z-axis is oriented vertically, so that the x and y axes lie in the horizontal plane.

Ordinarily, solid-state physics texts place the origin (0,0,0) of the perovskite unit cell at the barium site, with titanium in the center and the oxygens at the centers of the cube faces. Our choice of origin facilitates comparison with the structures of the oxide superconductors.

This structure is best understood in terms of the sizes of the atoms involved. The ionic radii of O^{2-} (1.32 Å) and Ba^{2+} (1.34 Å) are almost the same, as indicated in Table 7.1, and together they form a perfect fcc lattice with the smaller Ti^{4+} ions (0.68 Å) located in octahedral holes surrounded entirely by oxygens. The octahedral holes of a close-packed oxygen lattice have a radius of 0.545 Å; if these holes were empty the lattice constant would be $a = 3.73$ Å, as noted in Fig. 7.2a. Each titanium pushes the surrounding oxygens outward, as shown in Fig. 7.2b, thereby increasing the lattice constant. When the titanium is replaced by a larger atom, the lattice constant expands further, as indicated by the data in the last column of Table 7.2. When Ba is replaced by the smaller Ca (0.99 Å) and Sr (1.12 Å) ions, by contrast, there is a corresponding decrease in the lattice constant, as indicated by the data in columns 3 and 4, respectively, of Table 7.2. All three alkaline earths, Ca, Sr, and Ba, appear prominently in the structures of 3 high-temperature superconductors.

B. Tetragonal Form

At room temperature barium titanate is tetragonal and the deviation from cubic, $(c-a)/\frac{1}{2}(c+a)$, is about 1%. All of the atoms have the same x, y coordinates as in the cubic case, but are shifted along the z-axis relative to each other by ≈ 0.1 Å, producing the puckered arrangement shown in Fig. 7.3. The distortions from the ideal structure are exaggerated in this sketch. The puckering bends the Ti–O–Ti group so that the Ti–O distance increases while the Ti–Ti distance remains almost

Table 7.1 Ionic Radii for Selected Elements[a]

Small	Cu^{2+}	0.72 Å	Bi^{5+}	0.74 Å
Small–Medium	Cu^{+}	0.96 Å	Y^{3+}	0.94 Å
	Bi^{3+}	0.96 Å	Tl^{3+}	0.95 Å
	Ca^{2+}	0.99 Å	Bi^{3+}	0.96 Å
	Nd^{3+}	0.995 Å		
Medium–Large	Hg^{2+}	1.10 Å		
	Sr^{2+}	1.12 Å	La^{3+}	1.14 Å
	Pb^{2+}	1.20 Å	Ag^{+}	1.26 Å
Large	K^{+}	1.33 Å	O^{2-}	1.32 Å
	Ba^{2+}	1.34 Å	F^{-}	1.33 Å

[a] See Table VI-2 of Poole *et al.* (1988) for a more extensive list.

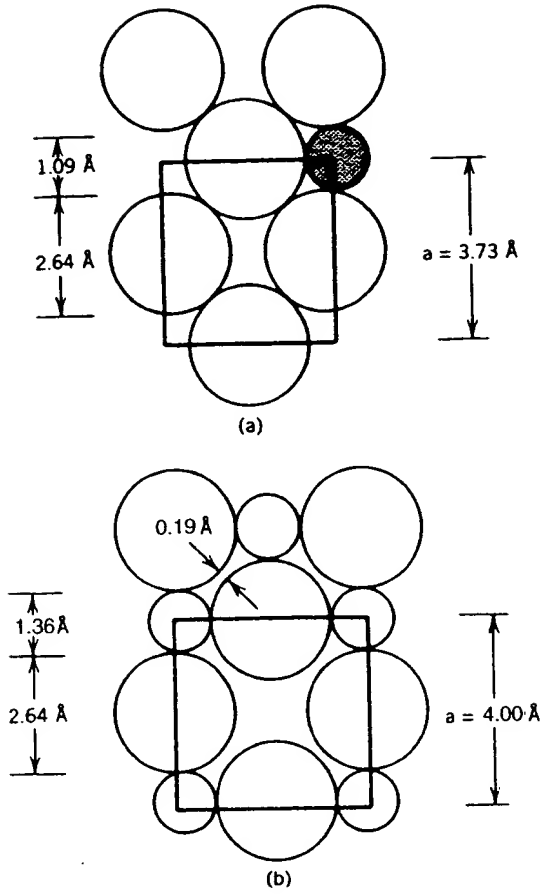


Figure 7.2 Cross section of the perovskite unit cell in the $z = 0$ plane showing (a) the size of the octahedral hole (shaded) between oxygens (large circles), and (b) oxygens pushed apart by the transition ions (small circles) in the hole sites. For each case the lattice constant is indicated on the right and the oxygen and hole sizes on the left (Poole *et al.*, 1988, p. 77).

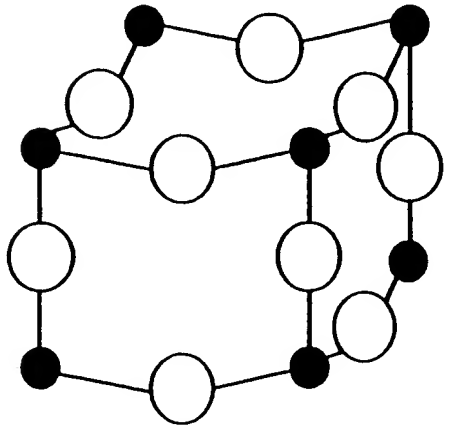


Figure 7.3 Perovskite tetragonal unit cell showing puckering of Ti–O layers that are perfectly flat in the cubic cell of Fig. 7.1. The notation of Fig. 7.1 is used (Poole *et al.*, 1988, p. 75).

the same. This has the effect of providing more room for the titanium atoms to fit in their lattice sites. We will see later that a similar puckering distortion occurs in the high-temperature superconductors as a way of providing space for the Cu atoms in the planes.

C. Orthorhombic Form

There are two principal ways in which a tetragonal structure distorts to form an orthorhombic phase. The first, shown at

Table 7.2 Dependence of Lattice Constants a of Selected Perovskites AMO_3 on Alkaline Earth A and Ionic Radius of Transition Metal Ion M^{+4}; the Alkaline Earth Ionic Radii are 0.99 Å (Ca), 1.12 Å (Sr), and 1.34 Å (Ba)[a]

Transitional metal	Transitional metal radius, Å	Lattice constant a, Å		
		Ca	Sr	Ba
Ti	0.68	3.84	3.91	4.01
Fe	—	—	3.87	4.01
Mo	0.70	—	3.98	4.04
Sn	0.71	3.92	4.03	4.12
Zr	0.79	4.02	4.10	4.19
Pb	0.84	—	—	4.27
Ce	0.94	3.85	4.27	4.40
Th	1.02	4.37	4.42	4.80

[a] Data from Wyckoff (1964, pp. 391ff).

the top of Fig. 7.4, is for the b-axis to stretch relative to the a-axis, resulting in the formation of a rectangle. The second, shown at the bottom of the figure, is for one diagonal of the ab square to stretch and the other diagonal to compress, resulting in the formation of a rhombus. The two diagonals are perpendicular, rotated by 45° relative to the original axes, and become the a', b' dimensions of the new orthorhombic unit cell, as shown in Fig. 7.5. These a', b' lattice constants are $\approx \sqrt{2}$ times longer than the original constants, so that the volume of the unit cell roughly doubles; thus, it contains exactly twice as many atoms. (The same $\sqrt{2}$ factor appears in Eq. 7.1 in our discussion of the lattice constants for the orthorhombic form of barium titanate.)

When barium titanate is cooled below 5°C it undergoes a diagonal- or rhombal-type distortion. The atoms have the same z coordinates ($z = 0$ or $\frac{1}{2}$) as in the cubic phase, so the distortion occurs entirely in the x, y-plane, with no puckering of the atoms. The deviation from tetragonality, as

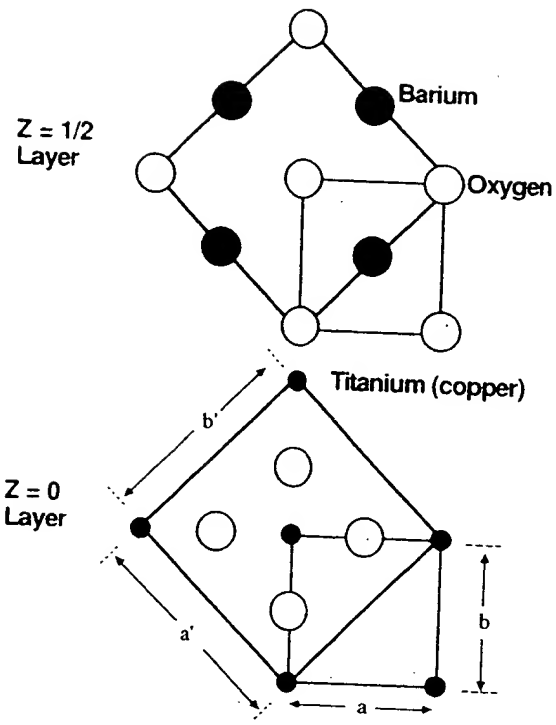


Figure 7.5 Rhombal expansion of monomolecular tetragonal unit cell (small squares, lower right) to bimolecular orthorhombic unit cell (large squares) with new axes 45° relative to the old axes. The atom positions are shown for the $z = 0$ and $z = \frac{1}{2}$ layers (Poole *et al.*, 1988, p. 76).

given by the percentage of anisotropy,

$$\% \text{ ANIS} = \frac{100|b - a|}{\frac{1}{2}(b + a)} = 0.22\%, \quad (7.3)$$

is less than that of most orthorhombic copper oxide superconductors. We see from Fig. 7.5 that in the cubic phase the oxygen atoms in the $z = 0$ plane are separated by 0.19 Å. The rhombal distortion increases this O–O separation in one direction and decreases it in the other, in the manner indicated in Fig. 7.6a, to produce the Ti nearest-neighbor configuration shown in Fig. 7.6b. This arrangement helps to fit the titanium into its lattice site.

The transformation from tetragonal to orthorhombic is generally of the rhombal type for $(La_{1-x}Sr_x)_2CuO_4$ and of the rectilinear type for $YBa_2Cu_3O_{7-\delta}$.

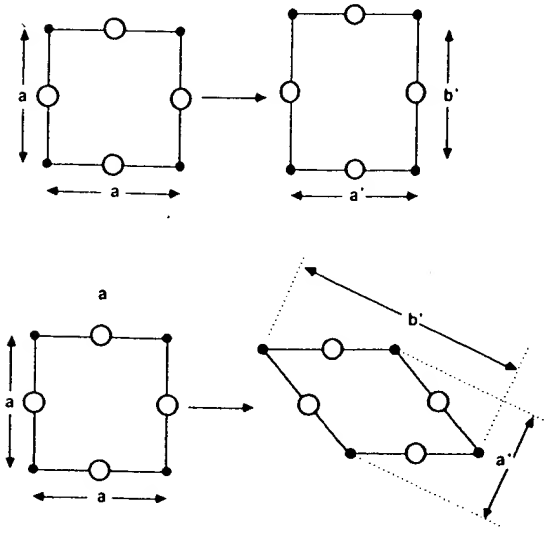


Figure 7.4 Rectangular- (top) and rhombal- (bottom) type distortions of a two-dimensional square unit cell of width a (Poole *et al.*, 1989).

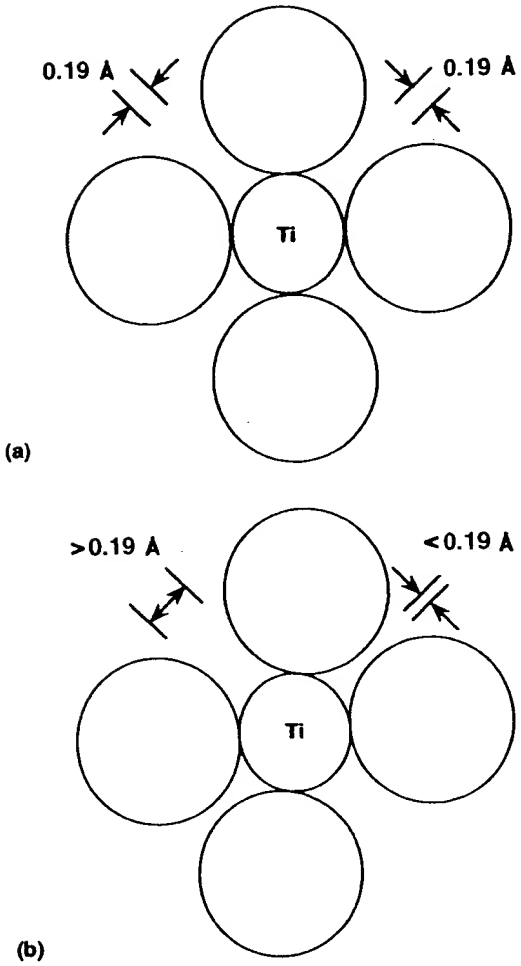


Figure 7.6 Shift of the oxygens in the a, b-plane around the titanium atom of perovskite from the room-temperature tetragonal (and cubic) configuration (a) to the rhombal configuration (b) of its low-temperature orthorhombic structure.

D. Planar Representation

Another way of picturing the structure of perovskite is to think of the atoms as forming horizontal planes. If we adopt the notation [E F C] to designate the occupation of the E, F, and C sites, the sketches of perovskite presented in Figs. 7.1 and 7.3 follow the scheme

$$\begin{array}{lll} z = 1 & [TiO_2-] & \text{Ti at E, O at two F sites} \\ z = \frac{1}{2} & [O-Ba] & \text{O at E, Ba at C} \\ z = 0 & [TiO_2-] & \text{Ti at E, O at two F sites.} \end{array} \qquad (7.4)$$

The planes at the heights $z = 0$, $\frac{1}{2}$, and 1 can be labeled using this notation. The usefulness of this labeling scheme will be clarified in Section V.

This completes our treatment of the structure of perovskite. We encountered many features that we will meet again in the analogous superconductor cases, and established notation that will be useful in describing the structure of the cuprates. However, before proceeding we present details about a cubic and a close-to-cubic perovskite superconductor in the following two sections.

III. CUBIC BARIUM POTASSIUM BISMUTH OXIDE

The compound

$$Ba_{1-x}K_xBiO_{3-y},$$

which forms for $x > 0.25$, crystallizes in the cubic pervoskite structure with $a = 4.29$ Å (Cava *et al.*, 1988; Jin *et al.*, 1992; Mattheiss *et al.*, 1988). K^+ ions replace some of the Ba^{2+} ions in the C site, and Bi ions occupy the E sites of Eq. (7.2) (Hinks *et al.*, 1988b; Kwei *et al.*, 1989; Pei *et al.*, 1990; Salem-Sugui *et al.*, 1991; Schneemeyer *et al.*, 1988). Some oxygen sites are vacant, as indicated by y. Hinks *et al.* (1989) and Pei *et al.* (1990) determined the structural phase diagram (cf. Kuentzler *et al.*, 1991; Zubkus *et al.*, 1991). We should note from Table 7.1 that the potassium (1.33 Å) and barium (1.32 Å) ions are almost the same size, and that Bi^{5+} (0.74 Å) is close to Ti^{4+} (0.68 Å). Bismuth represents a mixture of the valence states Bi^{3+} and Bi^{5+} which share the Ti^{4+} site in a proportion that depends on x and y. The larger size (0.96 Å) of the Bi^{3+} ion causes the lattice constant a to expand 7% beyond its cubic $BaTiO_3$ value. Oxygen vacancies help to compensate for the larger size of Bi^{3+}.

It is noteworthy that $Ba_{1-x}K_xBiO_{3-y}$ becomes superconducting at a temperature (≈ 40 K for $x \approx 0.4$) that is higher than the T_c of all of the $A15$ compounds. This compound, which has no copper, has

been widely studied in the quest for clues that would elucidate the mechanism of high-temperature superconductivity. Features of $Ba_{1-x}K_xBiO_{3-y}$, such as the fact that it contains a variable valence state ion and utilizes oxygen vacancies to achieve charge compensation, reappear in the high-temperature superconducting compounds.

IV. BARIUM LEAD BISMUTH OXIDE

In 1983 Mattheiss and Hamann referred to the 1975 "discovery by Sleight *et al.* of high-temperature superconductivity" in the compound $BaPb_{1-x}Bi_xO_3$ in the composition range $0.05 \leq x \leq 0.3$ with T_c up to 13 K. Many consider this system, which disproportionates $2Bi^{4+} \rightarrow Bi^{3+} + Bi^{5+}$ in going from the metallic to the semiconducting state, as a predecessor to the LaSrCuO system.

The metallic compound $BaPbO_3$ is a cubic perovskite with the relatively large lattice constant (Wyckoff, 1964; cf. Nitta *et al.*, 1965; Shannon and Bierstedt, 1970) listed in Table 7.3. At room temperature semiconducting $BaBiO_3$ is monoclinic ($a \approx b \approx c/\sqrt{2}$, $\beta = 90.17°$), but close to orthorhombic (Chaillout *et al.*, 1985; Cox and Sleight, 1976, 1979; cf. Federici *et al.*, 1990; Jeon *et al.*, 1990; Shen *et al.*, 1989). These two compounds form a solid solution series $BaPb_{1-x}Bi_xO_3$ involving cubic, tetragonal, orthorhombic, and monoclinic modifications. Superconductivity appears in the tetragonal phase, and the metal-to-insulator transition occurs at the tetragonal-to-orthorhombic phase boundary $x \approx 0.35$ (Gilbert *et al.*, 1978; Koyama and Ishimaru, 1992; Mattheiss, 1990; Mattheiss and Hamann, 1983; Sleight, 1987; cf. Bansil *et al.*, 1991; Ekino and Akimitsu, 1989a, b; Papaconstantopoulous *et al.*, 1989).

The compound resembles

$$Ba_{1-x}K_xBiO_{3-y}$$

with its variable Bi valence states, but it differs in not exhibiting superconductivity in the cubic phase.

V. PEROVSKITE-TYPE SUPERCONDUCTING STRUCTURES

In their first report on high-temperature superconductors Bednorz and Müller (1986) referred to their samples as "metallic, oxygen-deficient ... perovskite-like mixed-valence copper compounds." Subsequent work has confirmed that the new superconductors do indeed possess these characteristics.

In the oxide superconductors Cu^{2+} replaces the Ti^{4+} of perovskite, and in most cases the TiO_2-perovskite layering is retained as a CuO_2 layering with two oxygens per copper. Because of this feature of CuO_2 layers, which is common to all of the high-temperature superconductors, such superconductors exhibit a uniform lattice size in the a,b-plane, as the data in Table 7.3 demonstrate. The compound $BaCuO_3$ does not occur because the Cu^{4+} ion does not form, but this valence constraint is overcome by replacement of Ba^{2+} by a trivalent ion, such as La^{3+} or Y^{3+}, by a reduction in the oxygen content, or by both. The result is a set of "layers" containing only one oxygen per cation located between each pair of CuO_2 layers, or none at all. Each high-temperature superconductor has a unique sequence of layers.

We saw from Eq. (7.2) that each atom in perovskite is located in one of three types of sites. In like manner, each atom at the height z in a high-temperature superconductor occupies either an Edge (E) site on the edge $(0,0,z)$, a Face (F) site on the midline of a face $((0,\frac{1}{2},z)$ or $(\frac{1}{2},0,z)$ or both), or a Centered (C) site centered within the unit cell on the z-axis $(\frac{1}{2},\frac{1}{2},z)$. The site occupancy notation [E F C] is used because many cuprates contain a succession of [Cu O_2 −] and [− O_2 Cu] layers in which the Cu atom switches between edge and centered sites, with the oxygens

Table 7.3 Crystallographic Characteristics of Oxide Superconducting and Related Compounds[a]

Compound	Symbol	Symm	Type	Enlarg.	Form. units	a_0(Å)	c_0(Å)	c_0 / Cu	%Anis	T_c (K)	Comments
$BaTiO_3$	—	C	A	1	1	4.012	4.012	—	0	—	$T > 200°C$
$BaTiO_3$	—	T	A	1	1	3.995	4.03	—	0	—	20°C
$BaTiO_3$	—	O	A	$\sqrt{2}$	2	$4.013\sqrt{2}$	3.990	—	0.23	—	$T < 5°C$
$BaPbO_3$	—	C	A	1	1	4.273	4.273	—	0	0.4	
$BaPb_{0.7}Bi_{0.3}O_3$	—	T	S	$\sqrt{2}$	4	$4.286\sqrt{2}$	4.304	—	0	12	
$BaBiO_3$	—	M	A	$\sqrt{2}$	2	$4.355\sqrt{2}$	4.335	—	0.13	—	$\beta = 90.17°$
$Ba_{0.6}K_{0.4}BiO_3$	—	C	A	1	1	4.293	4.293	—	0	30	
La_2CuO_4	0201	T	S	1	2	3.81	13.18	6.59	0	35	Sr, doped
La_2CuO_4	0201	O	S	$\sqrt{2}$	4	$3.960\sqrt{2}$	13.18	6.59	6.85	35	Sr, doped
$YBa_2Cu_3O_8$	0213	T	A	1	1	3.902	11.94	3.98	0	—	
$YBa_2Cu_3O_7$	0213	O	A	1	1	3.855	11.68	3.89	1.43	92	
$Bi_2Sr_2CaCu_2O_8$	2212	T	S	$5\sqrt{2}$	20	$3.81\sqrt{2}$	30.6	7.65	0	84	
$Bi_2Sr_2Ca_2Cu_3O_{10}$	2223	O	S	$5\sqrt{2}$	20	$3.83\sqrt{2}$	37	6.17	0.57	110	
$Tl_2Ba_2CuO_6$	2201	T	S	1	2	3.83	23.24	11.6	0	90	
$Tl_2Ba_2CaCu_2O_8$	2212	T	S	1	2	3.85	29.4	7.35	0	110	
$Tl_2Ba_2Ca_2Cu_3O_{10}$	2223	T	S	1	2	3.85	35.88	5.98		125	
$TlBa_2CuO_5$	1201		A	1	1		9.5	9.5		< 17	
$TlBa_2CaCu_2O_7$	1212		A	1	1		12.7	6.35		91	
$TlBa_2Ca_2Cu_3O_9$	1223		A	1	1		15.9	5.3		116	
$TlBa_2Ca_3Cu_4O_{11}$	1234		A	1	1		19.1	4.78		122	
$TlBa_2Ca_4Cu_5O_{13}$	1245		A	1	1		22.3	4.46		< 120	
$HgBa_2CuO_4$	1201	T	A	1	1	3.86	9.5	9.5		95	
$HgBa_2CaCu_2O_6$	1212	T	A	1	1	3.86	12.6	6.3		122	
$HgBa_2Ca_2Cu_3O_8$	1223	T	A	1	1	3.86	17.7	5.2		133	

[a] Symbol, symmetry (cubic C, tetragonal T, orthorhombic O, monoclinic M); type (aligned A, staggered S); enlargement in a, b-plane (diagonal distortion $\sqrt{2}$, superlattice 5); formula units per unit cell; lattice parameters (a_0, c_0, c_0, and c_0 per Cu ion); % anisotropy; and transition temperature T_c. For the orthorhombic compounds tabulated values of a_0 are averages of a_0 and b_0.

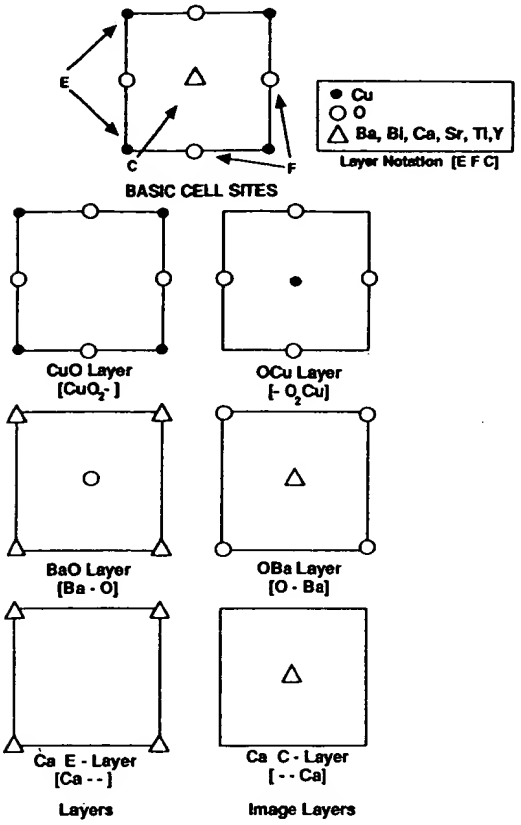


Figure 7.7 Types of atom positions in the layers of a high-temperature superconductor structure, using the edge, face, center notation [*E F C*]. Typical site occupancies are given in the upper right (Poole *et al.*, 1989).

remaining at their face positions. Similar alternations in position take place with Ba, O, and Ca layers, as illustrated in Fig. 7.7.

Hauck *et al.* (1991) proposed a classification of superconducting oxide structures in terms of the sequence (1) superconducting layers [Cu O_2 −] and [− O_2 Cu], (2) insulating layers, such as [Y − −] or [− − Ca], and (3) hole-donating layers, such as [Cu O^b −] or [Bi − O].

The high-temperature superconductor compounds have a horizontal reflection plane ($\perp$ to z) called σ_h at the center of the unit cell and another σ_h reflection plane at the top (and bottom). This means that every plane of atoms in the lower half of the cell at the height z is duplicated in the upper half at the height $1-z$. Such atoms, of course, appear twice in the unit cell, while atoms right on the symmetry planes only occur once since they cannot be reflected. Figure 7.8 shows a [Cu O_2 −] plane at a height z reflected to the height $1-z$. Note how the puckering preserves the reflection symmetry operation. Superconductors that have this reflection plane, but lack end-centering and body-centering op-

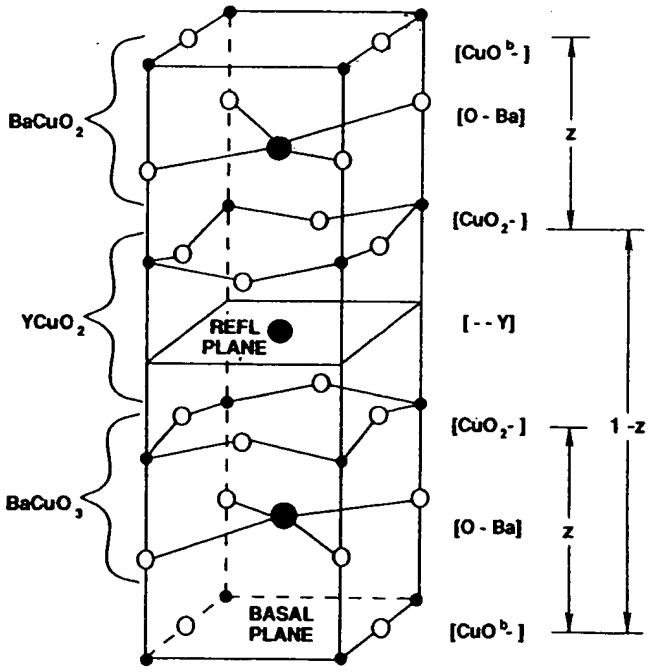


Figure 7.8 Unit cell of $YBa_2Cu_3O_7$ showing the molecular groupings, reflection plane, and layer types.

erations (see Section VII), are called *aligned* because all of their copper atoms are of one type; either all on the edge $(0,0,z)$ in E positions or all centered $(\frac{1}{2},\frac{1}{2},z)$ at C sites. In other words, they all lie one above the other on the same vertical lines, as do the Cu ions in Fig. 7.8.

VI. ALIGNED $YBa_2Cu_3O_7$

The compound $YBa_2Cu_3O_7$, sometimes called YBaCuO or the 123 compound, in its orthorhombic form is a superconductor below the transition temperature $T_c \approx 92$ K. Figure 7.8 sketches the locations of the atoms, Fig. 7.9 shows the arrangement of the copper oxide planes, Fig. 7.10 provides more details on the unit cell, and Table 7.4 lists the atom positions and unit cell dimensions (Beno *et al.*, 1987; Capponi *et al.*, 1987; Hazen *et al.*, 1987; Jorgensen *et al.*, 1987; Le Page *et al.*, 1987; Siegrist *et al.*, 1987; Yan and Blanchin, 1991; see also Schuller *et al.*, 1987). Considered as a perovskite derivative, it can be looked upon as a stacking of three perovskite units $BaCuO_3$, $YCuO_2$, and $BaCuO_2$, two of them with a missing oxygen, and this explains why $c \approx 3a$. It is, however, more useful to discuss the compound from the viewpoint of its planar structure.

A. Copper Oxide Planes

We see from Fig. 7.9 that three planes containing Cu and O are sandwiched between two planes containing Ba and O and one plane containing Y. The layering scheme is given on the right side of Fig. 7.8, where the superscript b on O indicates that the oxygen lies along the b-axis, as shown. The atoms are puckered in the two [Cu O_2 −] planes that have the [− − Y] plane between them. The third copper oxide plane [Cu O^b −], often referred to as

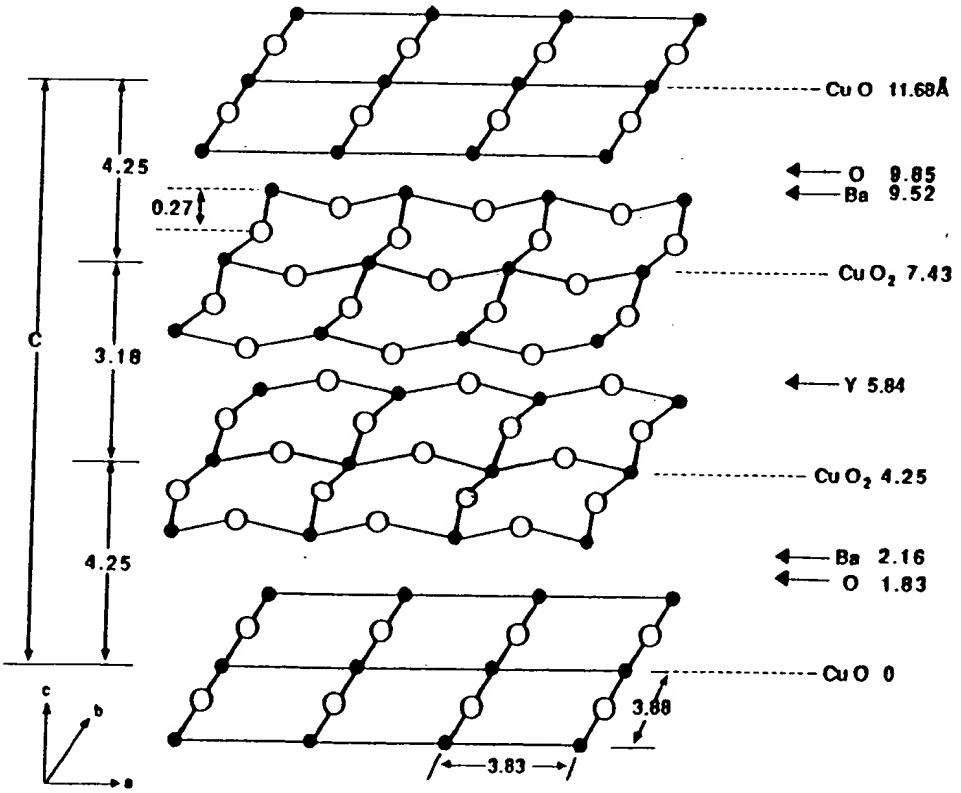


Figure 7.9 Layering scheme of orthorhombic $YBa_2Cu_3O_7$ with the puckering indicated. The layers are perpendicular to the c-axis (Poole *et al.*, 1988, p. 101).

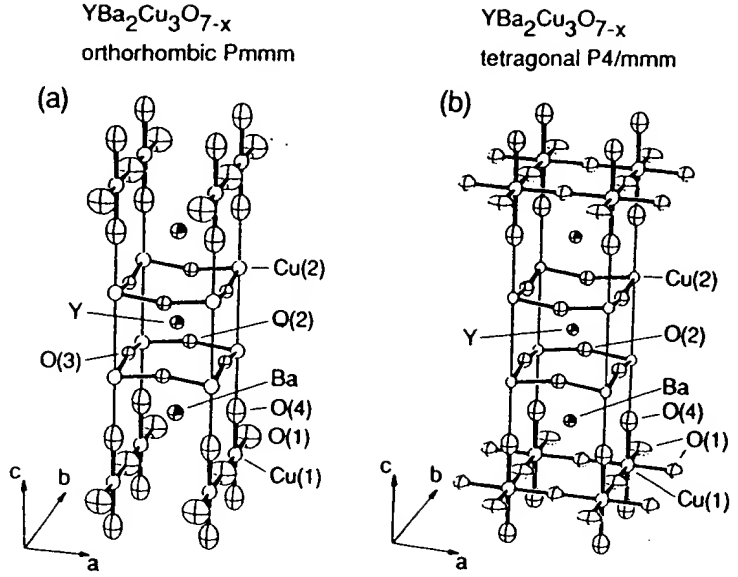


Figure 7.10 Sketches of the superconducting orthorhombic (left) and nonsuperconducting tetragonal (right) YBaCuO unit cells. Thermal vibration ellipsoids are shown for the atoms. In the tetragonal form the oxygen atoms are randomly dispersed over the basal plane sites (Jorgensen *et al.*, 1987a,b; also see Schuller *et al.*, 1987).

Table 7.4 Normalized Atom Positions in the $YBa_2Cu_3O_7$ Orthorhombic Unit Cell (dimensions $a = 3.83$ Å, $b = 3.88$ Å, and $c = 11.68$ Å)

Layer	Atom	x	y	z
[Cu O –]	Cu(1)	0	0	1
	O(1)	0	$\frac{1}{2}$	1
[O – Ba]	O(4)	0	0	0.8432
	Ba	$\frac{1}{2}$	$\frac{1}{2}$	0.8146
[Cu O_2 –]	Cu(2)	0	0	0.6445
	O(3)	0	$\frac{1}{2}$	0.6219
	O(2)	$\frac{1}{2}$	0	0.6210
[– – Y]	Y	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$
[Cu O_2 –]	O(2)	$\frac{1}{2}$	0	0.3790
	O(3)	0	$\frac{1}{2}$	0.3781
	Cu(2)	0	0	0.3555
[O – Ba]	Ba	$\frac{1}{2}$	$\frac{1}{2}$	0.1854
	O(4)	0	0	0.1568
[Cu O –]	O(1)	0	$\frac{1}{2}$	0
	Cu(1)	0	0	0

"the chains," consists of –Cu–O–Cu–O– chains along the *b* axis in lines that are perfectly straight because they are in a horizontal reflection plane σ_h; where no puckering can occur. Note that, according to the figures, the copper atoms are all stacked one above the other on edge (E) sites, as expected for an aligned-type superconductor. Both the copper oxide planes and the chains contribute to the superconducting properties.

B. Copper Coordination

Now that we have described the planar structure of YBaCuO it will be instructive to examine the local environment of each copper ion. The chain copper ion Cu(1) is square planar-coordinated and the two coppers Cu(2) and Cu(3) in the plane exhibit fivefold pyramidal coordination, as indicated in Fig. 7.11. The ellipsoids at the atom positions of Fig. 7.10 provide a measure of the thermal vibrational motion which the atoms experience, since the amplitudes of the atomic vibrations are indicated by the relative size of each of the ellipsoids.

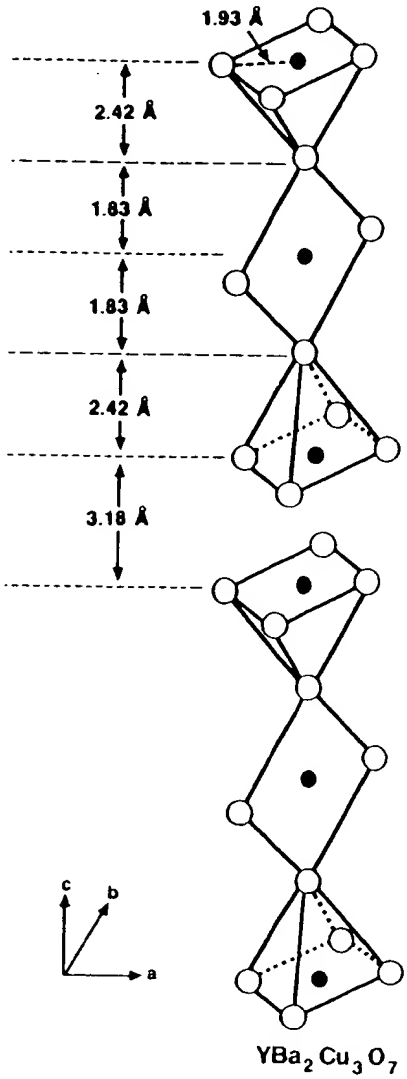


Figure 7.11 Stacking of pyramid, square–planar, and inverted pyramid groups along the *c*-axis of orthorhombic $YBa_2Cu_3O_7$ (adapted from Poole *et al.*, 1988, p. 100).

C. Stacking Rules

The atoms arrange themselves in the various planes in such a way as to enable them to stack one above the other in an efficient manner, with very little interference from neighboring atoms. Steric effects prevent large atoms such as Ba (1.34 Å) and O (1.32 Å) from overcrowding a layer or from aligning directly on top of each other in adjacent layers. In many cuprates stacking occurs in accordance with the following two empirical rules:

1. Metal ions occupy either edge or centered sites, and in adjacent layers alternate between E and C sites.
2. Oxygens are found in any type of site, but they occupy only one type in a particular layer, and in adjacent layers they are on different types of sites.

Minor adjustments to make more room can be brought about by puckering or by distorting from tetragonal to orthorhombic.

D. Crystallographic Phases

The $YBa_2Cu_3O_{7-\delta}$ compound comes in tetragonal and orthorhombic varieties, as shown in Fig. 7.10, and it is the latter phase which is ordinarily superconducting. In the tetragonal phase the oxygen sites in the chain layer are about half occupied

in a random or disordered manner, and in the orthorhombic phase are ordered into –Cu–O– chains along the b direction. The oxygen vacancy along the a direction causes the unit cell to compress slightly so that $a < b$, and the resulting distortion is of the rectangular type shown in Fig. 7.4a. Increasing the oxygen content so that $\delta < 0$ causes oxygens to begin occupying the vacant sites along a. Superlattice ordering of the chains is responsible for the phase that goes superconducting at 60 K.

YBaCuO is prepared by heating in the 750–900°C range in the presence of various concentrations of oxygen. The compound is tetragonal at the highest temperatures, increases its oxygen content through oxygen uptake and diffusion (Rothman *et al.*, 1991) as the temperature is lowered, and undergoes a second-order phase transition of the order–disorder type at about 700°C to the low-temperature orthorhombic phase, as indicated in Fig. 7.12

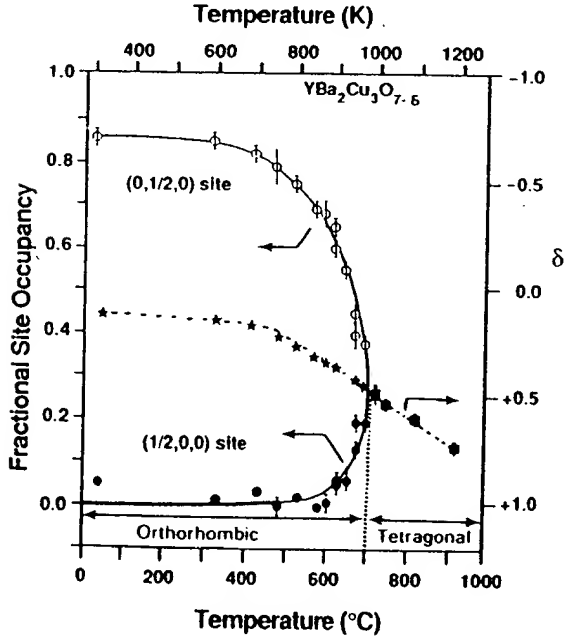


Figure 7.12 Fractional occupancies of the $(\frac{1}{2},0,0)$ (bottom) and $(0,\frac{1}{2},0)$ (top) sites (scale on left), and the oxygen content parameter δ (center, scale on right) for quench temperatures of YBaCuO in the range 0–1000°C. The δ parameter curve is the average of the two site-occupancy curves (adapted from Jorgensen *et al.*, 1987a; also see Schuller *et al.*, 1987; see also Poole *et al.*, 1988).

(Jorgensen *et al.*, 1987, 1990; Schuller *et al.*, 1987; cf. Beyers and Ahn, 1991; Metzger *et al.*, 1993; Fig. 8). Quenching by rapid cooling from a high temperature can produce at room temperature the tetragonal phase sketched on the right side of Fig. 7.10, and slow annealing favors the orthorhombic phase on the left. Figure 7.12 shows the fractional site occupancy of the oxygens in the chain site $(0,\frac{1}{2},0)$ as a function of the temperature in an oxygen atmosphere. A sample stored under sealed conditions exhibited no degradation in structure or change in T_c four years later (Sequeira *et al.*, 1992). Ultra-thin films tend to be tetragonal (Streiffer *et al.*, 1991).

E. Charge Distribution

Information on the charge distributions around atoms in conductors can be obtained from knowledge of their energy bands (see description in Chapter 8). This is most easily accomplished by carrying out a Fourier-type mathematical transformation between the reciprocal k_x, k_y, k_z-space (cf. Chapter 8, Section II) in which the energy bands are plotted and the coordinate x, y, z-space, where the charge is distributed. We will present the results obtained for $YBa_2Cu_3O_7$ in the three vertical symmetry planes (x,z, y,z, and diagonal), all containing the z-axis through the origin, shown shaded in the unit cell of Fig. 7.13.

Contour plots of the charge density of the valence electrons in these planes are sketched in Fig. 7.14. The high density at the Y^{3+} and Ba^{2+} sites and the lack of contours around these sites together indicate that these atoms are almost completely ionized, with charges of +3 and +2, respectively. It also shows that these ions are decoupled from the planes above and below. This accounts for the magnetic isolation of the Y site whereby magnetic ions substituted for yttrium do not interfere with the superconducting properties. In contrast, the contours surrounding the Cu and O ions are not characteristic of an

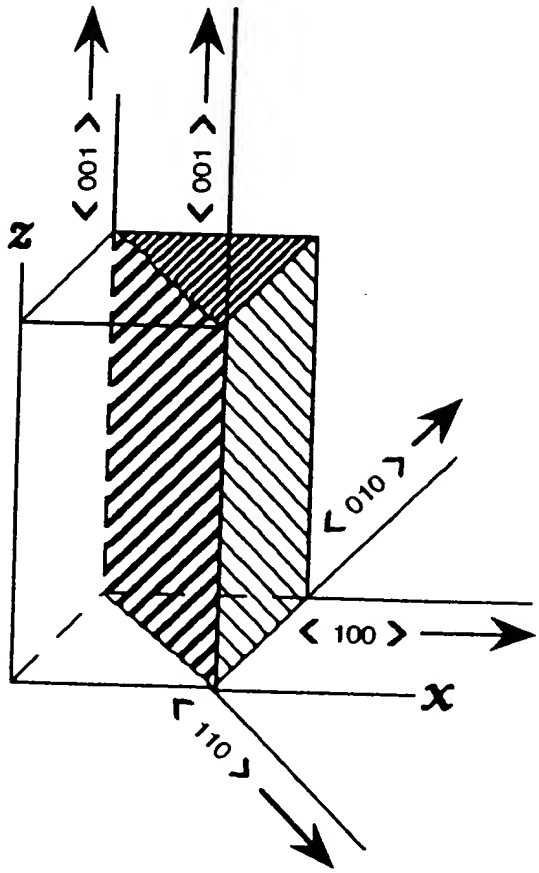


Figure 7.13 Three vertical crystallographic planes (x, z-, y, z-, and diagonal) of a tetragonal unit cell of $YBa_2Cu_3O_7$, and standard notation for the four crystallographic directions.

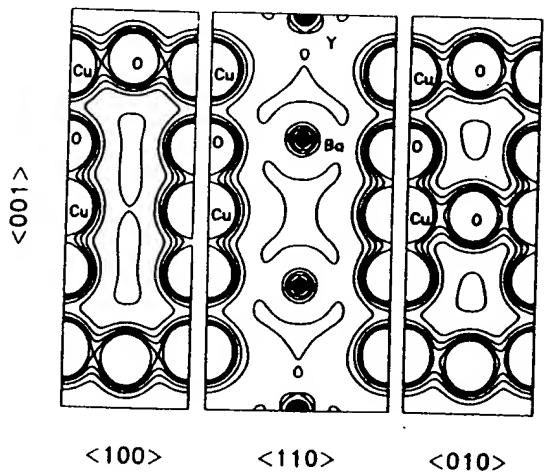


Figure 7.14 Charge density in the three symmetry planes of YBaCuO shown shaded in Fig. 7.13. The x, z, diagonal and the y, z planes are shown from left to right, labeled ⟨100⟩, ⟨110⟩, and ⟨010⟩, respectively. These results are obtained from band structure calculations, as will be explained in the following chapter (Krakauer and Pickett, 1988).

ordinary ionic compound. The short Cu–O bonds in the planes and chains (1.93–1.96 Å) increase the charge overlap. The least overlap appears in the Cu(2)–O(4) vertical bridging bond, which is also fairly long (2.29 Å). The Cu, O charge contours can be represented by a model that assigns charges of +1.62 and −1.69 to Cu and O, respectively, rather than the values of +2.33 and −2.00 expected for a standard ionic model, where the charge +2.33 is an average of +2, +2, and +3 for the three copper ions. Thus the Cu–O bonds are not completely ionic, but partly covalent.

F. YBaCuO Formula

In early work the formula

$$YBa_2Cu_3O_{9-\delta}$$

was used for YBaCuO because the prototype triple pervoskite $(YCuO_3)(BaCuO_3)_2$ has nine oxygens. Then crystallographers showed that there are eight oxygen sites in the 14-atom YBaCuO unit cell, and the formula $YBa_2Cu_3O_{8-\delta}$ came into widespread use. Finally, structure refinements demonstrated that one of the oxygen sites is systematically vacant in the chain layers, so the more appropriate expression $YBa_2Cu_3O_{7-\delta}$ was introduced. It would be preferable to make one more change and use the formula $Ba_2YCu_3O_{7-\delta}$ to emphasize that Y is analogous to Ca in the bismuth and thallium compounds, but very few workers in the field do this, so we reluctantly adopt the usual "final" notation. In the Bi–Tl compound notation of Section IX, B, $Ba_2YCu_3O_{7-\delta}$ would be called a 0213 compound. We will follow the usual practice of referring to $YBa_2Cu_3O_{7-\delta}$ as the 123 compound.

G. $YBa_2Cu_4O_8$ and $Y_2Ba_4Cu_7O_{15}$

These two superconductors are sometimes referred to as the 124 compound and the 247 compound, respectively. They have the property that for each atom at position (x, y, z) there is another identical atom at

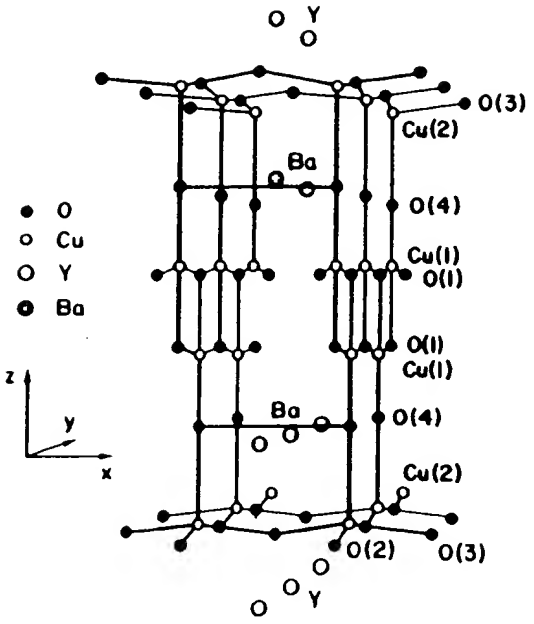


Figure 7.15 Crystal structure of $YBa_2Cu_4O_8$ showing how, as a result of the side-centering symmetry operation, the atoms in adjacent Cu–O chains are staggered along the y direction, with Cu above O and O above Cu (Heyen *et al.*, 1991; modified from Campuzano *et al.*, 1990).

position $(x, y + \frac{1}{2}, z + \frac{1}{2})$. In other words, the structure is side centered. This property prevents the stacking rules of Section C from applying.

The chain layer of $YBa_2Cu_3O_7$ becomes two adjacent chain layers in $YBa_2Cu_4O_8$, with the Cu atoms of one chain located directly above or below the O atoms of the other, as shown in Fig. 7.15 (Campuzano *et al.*, 1990; Heyen *et al.*, 1990a, 1991; Iqbal, 1992; Kaldis *et al.*, 1989; Marsh *et al.*, 1988; Morris *et al.*, 1989a). The transition temperature remains in the range from 40 K to 80 K when Y is replaced by various rare earths (Morris *et al.*, 1989). The double chains do not exhibit the variable oxygen stoichiometry of the single ones.

The other side-centered compound, $Y_2Ba_4Cu_7O_{15}$, may be considered according to Torardi, "as an ordered 1:1 intergrowth of the 123 and 124 compounds

$$(YBa_2Cu_3O_7 + YBa_2Cu_4O_8 = Y_2Ba_4Cu_7O_{15})"$$

(Bordel *et al.*, 1988, Gupta and Gupta, 1993). The 123 single chains can vary in their oxygen content, and superconductivity onsets up to 90 K have been observed. This compound has been synthesized with several rare earths substituted for Y (Morris *et al.*, 1989b).

VII. BODY CENTERING

In Section V we discussed aligned-type superconductor structures that possess a horizontal plane of symmetry. Most high-temperature superconductor structures have, besides this σ_h plane, an additional symmetry operation called body centering whereby for every atom with coordinates (x, y, z) there is an identical atom with coordinates as determined from the following operation:

$$x \rightarrow x \pm \tfrac{1}{2}, \quad x \rightarrow y \pm \tfrac{1}{2}, \quad z \rightarrow z \pm \tfrac{1}{2} \quad (7.5)$$

Starting with a plane at the height z this operation forms what is called an image plane at the height $z + \frac{1}{2}$ in which the edge atoms become centered, the centered atoms become edge types, and each face atom moves to another face site. In other words, the body-centering operation acting on a plane at the height z forms a body centered plane, also called an image plane, at the height $z \pm \frac{1}{2}$. The signs in these operations are selected so that the generated points and planes remain within the unit cell. Thus if the initial value of z is greater than $\frac{1}{2}$, the minus sign must be selected, viz., $z \rightarrow z - \frac{1}{2}$. Body centering causes half of the Cu–O planes to be [Cu O_2 −], with the copper atoms at edge sites, and the other half to be [− O_2 Cu], with the copper atoms at centered sites.

Let us illustrate the symmetry features of a body-centered superconductor by considering the example of $Tl_2Ba_2CaCu_2O_8$. This compound has an initial plane [Cu O_2 −] with the copper and oxygen atoms at the vertical positions $z = 0.0540$ and 0.0531, respectively, as shown in Fig.

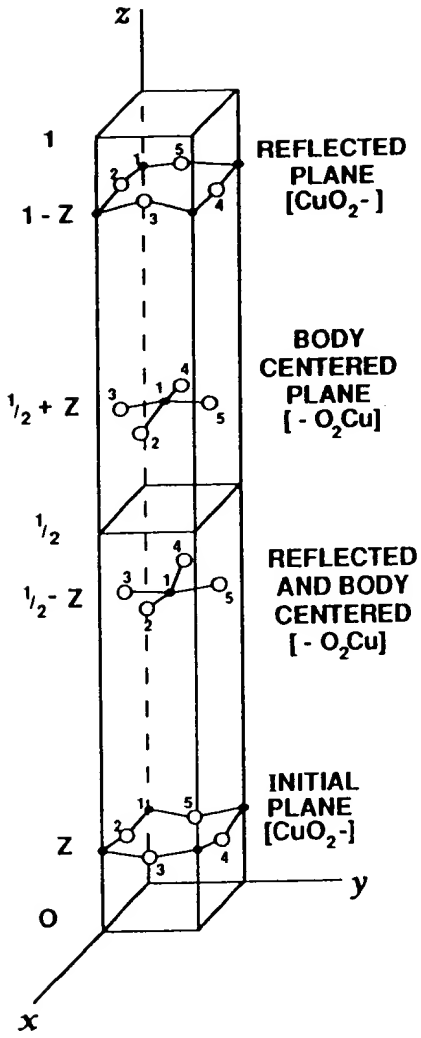


Figure 7.16 Body-centered tetragonal unit cell containing four puckered CuO_2 groups showing how the initial group (bottom) is replicated by reflection in the horizontal reflection plane ($z = \frac{1}{2}$), by the body centering operation, and by both.

7.16. For illustrative purposes the figure is drawn for values of z closer to 0.1. We see from the figure that there is a reflected plane [Cu O_2 −] at the height $1 - z$, an image (i.e., body centered) plane [− O_2 Cu] of the original plane at the height $\frac{1}{2} + z$, and an image plane [− O_2 Cu] of the reflected plane (i.e., a reflected and body centered plane) at the height $\frac{1}{2} - z$. Figure 7.16 illustrates this situation and indicates how the atoms of the initial plane can be transformed into particular atoms in other planes (see Problem 5). Figure 7.17 shows how the configurations of the

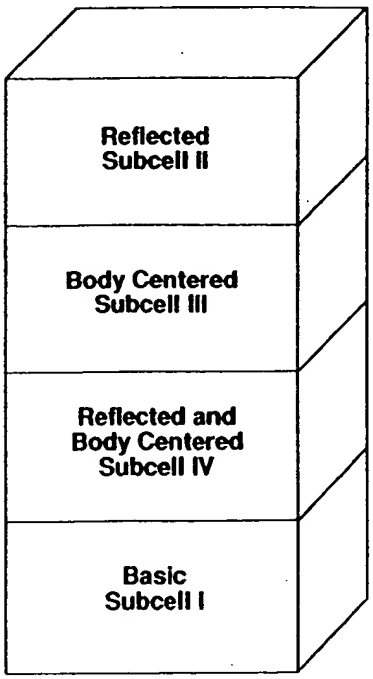


Figure 7.17 Body-centered unit cell divided into four regions by the reflection and body centering operations.

atoms in one-quarter of the unit cell, called the *basic subcell*, or subcell I, determine their configurations in the other three subcells II, III, and IV through the symmetry operations of reflection and body centering.

VIII. BODY-CENTERED La_2CuO_4 AND Nd_2CuO_4

The body-centered compound

$$M_2CuO_4$$

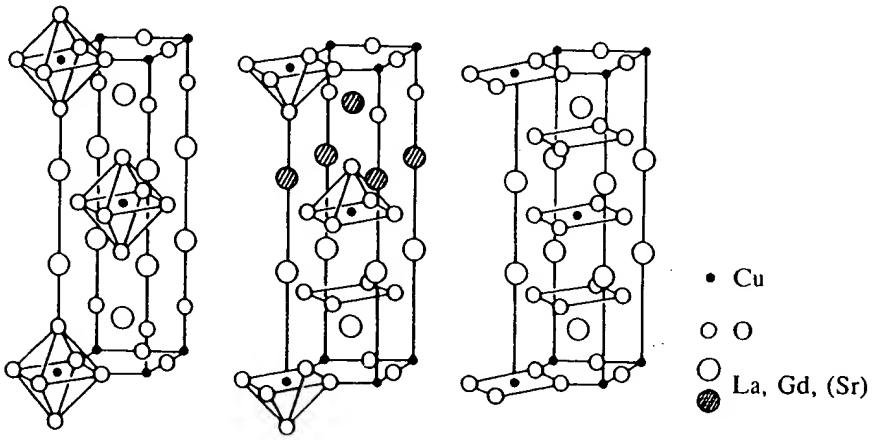
has three structural variations in the same crystallographic space group, namely the $M = La$ and $M = Nd$ types, and a third mixed variety (Xiao *et al.*, 1989). Table 7.5 lists the atom positions of the first two types, and Fig. 7.18 presents sketches of the structures of all three. Each will be discussed in turn.

A. Unit Cell Generation of La_2CuO_4 (T Phase)

The structure of the more common La_2CuO_4 variety, often called the T phase,

Table 7.5 Atom Positions in the La_2CuO_4 and Nd_2CuO_4 Structures

La_2CuO_4 structure					Nd_2CuO_4 structure				
Layer	**Atom**	x	y	z	**Layer**	**Atom**	x	y	z
	O(1)	$\frac{1}{2}$	0	1		O(1)	$\frac{1}{2}$	0	1
[Cu O_2 −]	Cu	0	0	1	[Cu O_2 −]	Cu	0	0	1
	O(1)	0	$\frac{1}{2}$	1		O(1)	0	$\frac{1}{2}$	1
	La	$\frac{1}{2}$	$\frac{1}{2}$	0.862	[− − Nd]	Nd	$\frac{1}{2}$	$\frac{1}{2}$	0.862
[O − La]									
	O(2)	0	0	0.818		O(3)	0	$\frac{1}{2}$	$\frac{3}{4}$
					[− O_2 −]				
	O(2)	$\frac{1}{2}$	$\frac{1}{2}$	0.682		O(3)	$\frac{1}{2}$	0	$\frac{3}{4}$
[La − O]									
	La	0	0	0.638	[Nd − −]	Nd	0	0	0.638
	O(1)	$\frac{1}{2}$	0	$\frac{1}{2}$		O(1)	$\frac{1}{2}$	0	$\frac{1}{2}$
[− O_2 Cu]	Cu	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	[− O_2 Cu]	Cu	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$
	O(1)	0	$\frac{1}{2}$	$\frac{1}{2}$		O(1)	0	$\frac{1}{2}$	$\frac{1}{2}$
	La	0	0	0.362	[Nd − −]	Nd	0	0	0.362
[La − O]									
	O(2)	$\frac{1}{2}$	$\frac{1}{2}$	0.318		O(3)	$\frac{1}{2}$	0	$\frac{1}{4}$
					[− O_2 −]				
	O(2)	0	0	0.182		O(3)	0	$\frac{1}{2}$	$\frac{1}{4}$
[O − La]									
	La	$\frac{1}{2}$	$\frac{1}{2}$	0.138	[− − Nd]	Nd	$\frac{1}{2}$	$\frac{1}{2}$	0.138
	O(1)	0	$\frac{1}{2}$	0		O(1)	0	$\frac{1}{2}$	0
[Cu O_2 −]	Cu	0	0	0	[Cu O_2 −]	Cu	0	0	0
	O(1)	$\frac{1}{2}$	0	0		O(1)	$\frac{1}{2}$	0	0



(a) T phase (b) T* phase (c) T′ phase

Figure 7.18 (a) Regular unit cell (T phase) associated with hole-type $(La_{1-x}Sr_x)_2CuO_4$ superconductors, (b) hybrid unit cell (T* phase) of the hole-type $La_{2-x-y}R_ySr_xCuO_4$ superconductors, and (c) alternate unit cell (T′ phase) associated with electron-type $(Nd_{1-x}Ce_x)_2CuO_4$ superconductors. The La atoms in the left structure become Nd atoms in the right structure. The upper part of the hybrid cell is T type, and the bottom is T′. The crystallographic space group is the same for all three unit cells (Xiao *et al.*, 1989; see also Oguchi, 1987; Ohbayashi *et al.*, 1987; Poole *et al.*, 1988, p. 83; Tan *et al.*, 1990).

can be pictured as a stacking of CuO_4La_2 groups alternately with image (i.e., body centered) La_2O_4Cu groups along the *c* direction, as indicated on the left side of Fig. 7.19 (Cavaet *et al.*, 1987; Kinoshita *et al.*, 1992; Longo and Raccah, 1973; Ohbayashi *et al.*, 1987; Onoda *et al.*, 1987; Zolliker *et al.*, 1990). Another way of visualizing the structure is by generating it from the group $Cu_{\frac{1}{2}}O_2La$, comprising the layers [O–La] and $\frac{1}{2}$[Cu O_2 –] in subcell I shown on the right side of Fig. 7.19 and also on the left side of Fig. 7.20. (The factor $\frac{1}{2}$ appears because the [Cu O_2 –] layer is shared by two subcells.) Subcell II is formed by reflection from subcell I, and subcells III and IV are formed from I and II via the body-centering operation in the manner of Figs. 7.16 and 7.17. Therefore, subcells I and II together contain the group CuO_4La_2, and subcells III and IV together contain its image (body centered) counterpart group La_2O_4Cu. The BiSrCaCuO and TlBaCaCuO structures to be discussed in Section IX can be generated in the same manner, but with much larger repeat units along the *c* direction.

B. Layering Scheme

The La_2CuO_4 layering scheme consists of equally-spaced, flat CuO_2 layers with their oxygens stacked one above the other, the copper ions alternating between the (0,0,0) and $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ sites in adjacent layers, as shown in Fig. 7.21. These planes are body-centered images of each other, and are perfectly flat because they are

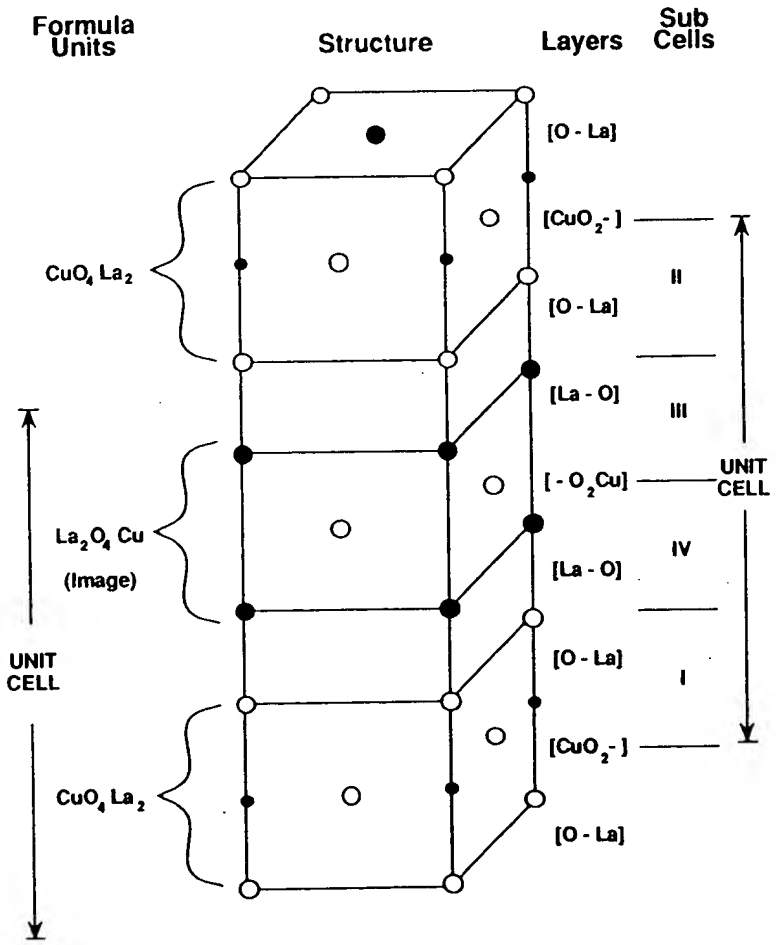


Figure 7.19 Structure of La_2CuO_4 (center), showing the formula units (left) and the level labels and subcell types (right). Two choices of unit cell are indicated, the left-side type unit cell based on formula units, and the more common right-side type unit cell based on copper-oxide layers.

La_2CuO_4	Sub Cell	Nd_2CuO_4
$[CuO_2-]$	——	$[CuO_2-]$
[O - La]	II	[- - Nd]
[La - O]	——	$[-O_2-]$
	III	[Nd - -]
$[-O_2Cu]$	——	$[-O_2Cu]$
[La - O]	IV	[Nd - -]
[O - La]	——	$[-O_2-]$
	I	[- - Nd]
$[CuO_2-]$	——	$[CuO_2-]$

Figure 7.20 Layering schemes of the La_2CuO_4 (T, left) and Nd_2CuO_4 (T', right) structures. The locations of the four subcells of the unit cell are indicated in the center column.

reflection planes. Half of the oxygens, O(1), are in the planes, and the other half, O(2), between the planes. The copper is octahedrally coordinated with oxygen, but the distance 1.9 Å from Cu to O(1) in the CuO_2 planes is much less than the vertical distance of 2.4 Å from Cu to the apical oxygen O(2), as indicated in Fig. 7.22. The La is ninefold coordinated to four O(1) oxygens, to four O(2) at $(\frac{1}{2}, \frac{1}{2}, z)$ sites, and to one O(2) at a $(0, 0, z)$ site.

C. Charge Distribution

Figure 7.23 shows contours of constant-valence charge density on a logarithmic scale drawn on the back x, z-plane and on the diagonal plane of the unit cell sketched in Fig. 7.13. These contour plots are obtained from the band structure calculations described in Chapter 8, Section XIV. The high-charge density at the lanthanum site and the low charge density around this site indicate an ionic state

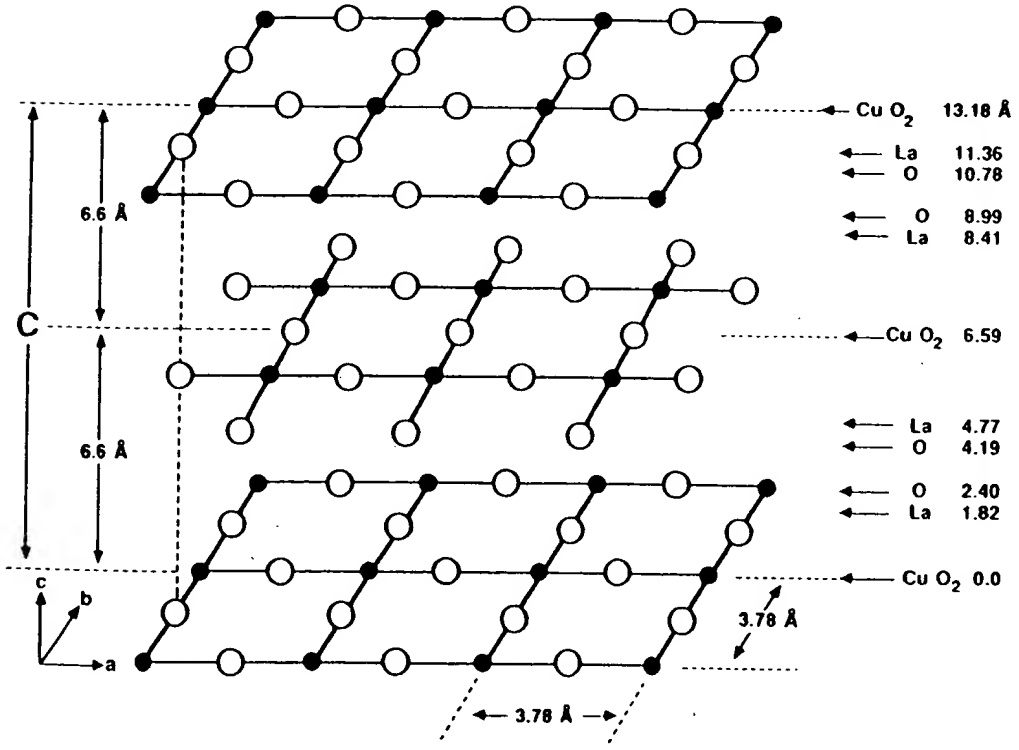


Figure 7.21 CuO_2 layers of the La_2CuO_4 structure showing horizontal displacement of Cu atoms in alternate layers. The layers are perpendicular to the c-axis (Poole *et al.*, 1988, p. 87).

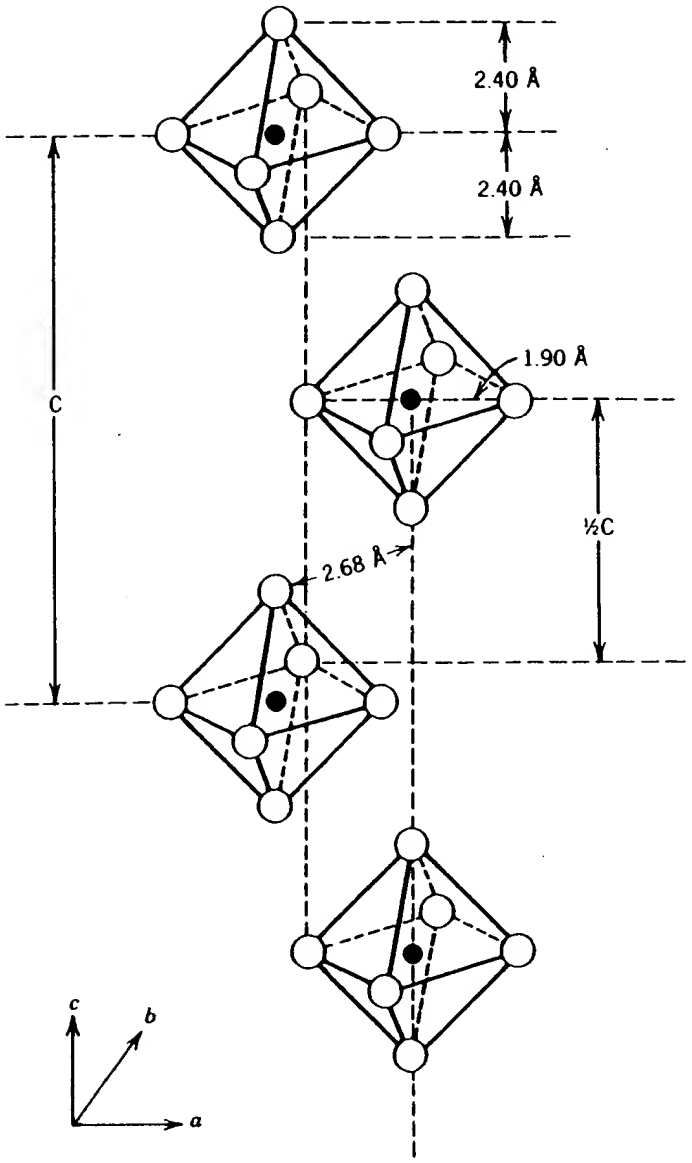


Figure 7.22 Ordering of axially distorted CuO_6 octahedra in La_2CuO_4 (Poole *et al.*, 1988, p. 88).

La^{3+}. The charge density changes in a fairly regular manner around the copper and oxygen atoms, both within the CuO_2 planes and perpendicular to these planes, suggestive of covalency in the Cu–O bonding, as is the case with the $YBa_2Cu_3O_7$ compound.

D. Superconducting Structures

The compound La_2CuO_4 is itself an antiferromagnetic insulator and must be doped, generally with an alkaline earth, to exhibit pronounced superconducting properties. The compounds $(La_{1-x}M_x)_2CuO_4$, with 3% to 15% of M = Sr or Ba replacing La, are orthorhombic at low temperatures and low M contents and are tetragonal otherwise; superconductivity has been found on both sides of this transition. The orthorhombic distortion can be of the rectangular or of the rhombal type, both of which are sketched in Fig. 7.4. The phase diagram of Fig. 7.24 shows the tetragonal, orthorhombic, superconducting, and antiferromagnetically ordered regions for the lanthanum compound (Weber *et al.*, 1989; cf. Goodenough *et al.*, 1993). We see that

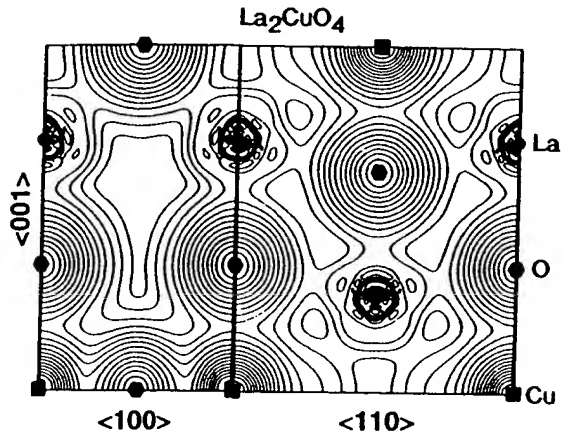


Figure 7.23 Contour plots of the charge density of La_2CuO_4 obtained from band structure calculations. The x, z-crystallographic plane labeled ⟨100⟩ is shown on the left and the diagonal plane labeled ⟨110⟩ on the right. The contour spacing is on a logarithmic scale (Pickett, 1989).

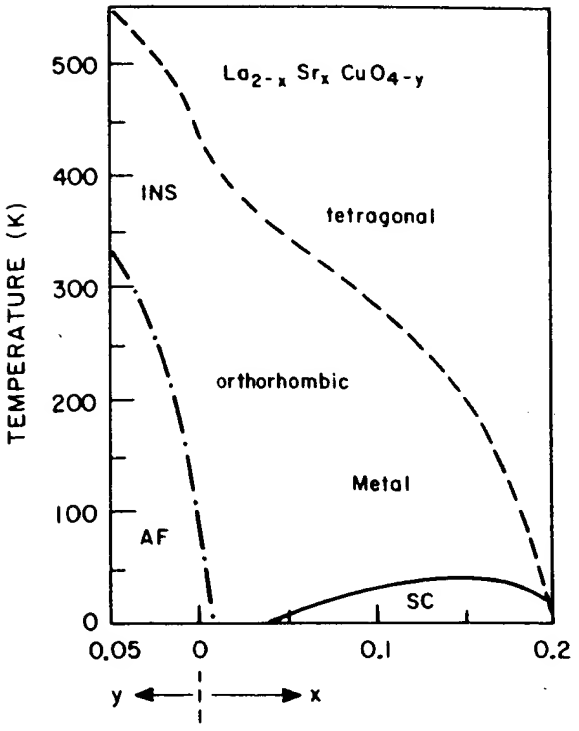


Figure 7.24 Phase diagram for hole-type $La_{2-x}Sr_xCuO_{4-y}$ indicating insulating (INS), antiferromagnetic (AF), and superconducting (SC) regions. Figure VI-6 of Poole *et al.* (1988) shows experimental data along the orthorhombic-to-tetragonal transition line. Spin-density waves (SDW) are found in the AF region (Weber *et al.*, 1989).

the orthorhombic phase is insulating at high temperatures, metallic at low temperatures, and superconducting at very low temperatures. Spin-density waves, to be discussed in Chapter 8, Section XIX, occur in the antiferromagnetic region.

E. Nd_2CuO_4 Compound (T′ Phase)

The rarer Nd_2CuO_4 structure (Skantakumar *et al.*, 1989; Sulewski *et al.*, 1990; Tan *et al.*, 1990) given on the right side of Fig. 7.18 and Table 7.5 has all of its atoms in the same positions as the standard La_2CuO_4 structure, except for the apical O(2) oxygens in the [O–La] and [La–O] layers, which move to form a [– O_2 –] layer between [– – La] and [La – –]. These oxygens, now called O(3), have the same x, y coordinate positions as the O(1) oxygens, and are located exactly between the CuO_2 planes with $z = \frac{1}{4}$ or $\frac{3}{4}$. We see from Fig. 7.18 that the CuO_6 octahedra have now lost their apical oxygens, causing Cu to become square planar-coordinated CuO_4 groups. The Nd is eightfold coordinated to four O(1) and four O(3) atoms, but with slightly different Nd–O distances. The CuO_2 planes, however, are identical in the two structures. Superconductors with this Nd_2CuO_4 structure are of the electron type, in contrast to other high-temperature superconductors, in which the current carriers are holes. In particular, the electron superconductor $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ with $T_c = 24$ K has been widely studied (Fontcuberta and Fàbrega, 1995, a review chapter; Allen 1990; Alp *et al.*, 1989b; Barlingay *et al.*, 1990; Ekino and Akimitsu, 1989a,b; Lederman *et al.*, 1991; Luke *et al.*, 1990; Lynn *et al.*, 1990; Sugiyama *et al.*, 1991; Tarason *et al.*, 1989a). Other rare earths, such as Pr (Lee *et al.*, 1990) and Sm (Almasan *et al.*, 1992) have replaced Nd.

The difference of structures associated with different signs attached to the current carriers may be understood in terms of the doping process that converts undoped material into a superconductor. Lanthanum and neodymium are both trivalent, and in the undoped compounds they each con-

tribute three electrons to the nearby oxygens,

$$\begin{aligned} \mathrm{La} &\rightarrow \mathrm{La}^{3+} + 3\mathrm{e}^-, \\ \mathrm{Nd} &\rightarrow \mathrm{Nd}^{3+} + 3\mathrm{e}^-, \end{aligned} \tag{7.6}$$

to produce O^{2-}. To form the superconductors a small amount of La in La_2CuO_4 can be replaced with divalent Sr, and some Nd in Nd_2CuO_4 can be replaced with tetravalent Ce, corresponding to

$$\begin{aligned} \mathrm{Sr} &\rightarrow \mathrm{Sr}^{2+} + 2\mathrm{e}^- \quad (\text{in } \mathrm{La_2CuO_4}) \\ \mathrm{Ce} &\rightarrow \mathrm{Ce}^{4+} + 4\mathrm{e}^- \quad (\text{in } \mathrm{Nd_2CuO_4}). \end{aligned} \tag{7.7}$$

Thus, Sr doping decreases the number of electrons to produce hole-type carriers, while Ce doping increases the electron concentration and the conductivity is electron type.

There are also copper-oxide electron superconductors with different structures, such as $Sr_{1-x}Nd_xCuO_2$ (Smith *et al.*, 1991) and $TlCa_{1-x}R_xSr_2Cu_2O_{7-\delta}$, where R is a rare earth (Vijayaraghavan *et al.*, 1989). Electron- and hole-type superconductivity in the cuprates has been compared (Katti and Risbud, 1992; Medina and Regueiro, 1990).

F. $La_{2-x-y}R_xSr_yCuO_4$ Compounds (T* Phase)

We have described the T structure of La_2CuO_4 and the T′ structure of Nd_2CuO_4. The former has O(2) oxygens and the latter O(3) oxygens, which changes the coordinations of the Cu atoms and that of the La and Nd atoms as well. There is a hybrid structure of hole-type superconducting lanthanum cuprates called the T* structure, illustrated in Fig. 7.18b, in which the upper half of the unit cell is the T type with O(2) oxygens and lower half the T′ type with O(3) oxygens. These two varieties of halfcells are stacked alternately along the tetragonal c-axis (Akimitsu *et al.*, 1988; Cheong *et al.*, 1989b; Kwei *et al.*, 1990; Tan *et al.*, 1990). Copper, located in the base of an oxygen pyramid, is fivefold-coordinated CuO_5. There are two inequivalent rare earth sites; the ninefold-coordinated site in the T-type halfcell is preferentially occupied by the larger La and Sr ions, while the smaller rare earths R (i.e., Sm, Eu, Gd, or Tb) prefer the eightfold-coordinated site in the T′ halfcell. Tan *et al.* (1991) give a phase diagram for the concentration ranges over which the T and T* phases are predominant.

IX. BODY-CENTERED BiSrCaCuO AND TlBaCaCuO

Early in 1988 two new superconducting systems with transition temperatures considerably above those attainable with YBaCuO, namely the bismuth- and thallium-based materials, were discovered. These compounds have about the same a and b lattice constants as the yttrium and lanthanum compounds, but with much larger unit cell dimensions along c. We will describe their body-centered structures in terms of their layering schemes. In the late 1940s some related compounds were synthesized by the Swedish chemist Bengt Aurivillius (1950, 1951, 1952).

A. Layering Scheme

The $Bi_2Sr_2Ca_nCu_{n+1}O_{6+2n}$ and $Tl_2Ba_2Ca_nCu_{n+1}O_{6+2n}$ compounds, where n is an integer, have essentially the same structure and the same layering arrangement (Barry *et al.*, 1989; Siegrist *et al.*, 1988; Torardi *et al.*, 1988a; Yvon and François, 1989), although there are some differences in the detailed atom positions. Here there are groupings of CuO_2 layers, each separated from the next by Ca layers with no oxygen. The CuO_2 groupings are bound together by intervening layers of BiO and SrO for the bismuth compound, and by intervening layers of TlO and BaO for the thallium compound. Figure 7.25 compares the layering scheme of the $Tl_2Ba_2Ca_nCu_{n+1}O_{6+2n}$ compounds with $n = 0, 1, 2$ with those of the lanthanum and yttrium compounds. We also see from the figure that the groupings of [Cu O_2 −] planes and [− O_2 Cu] image

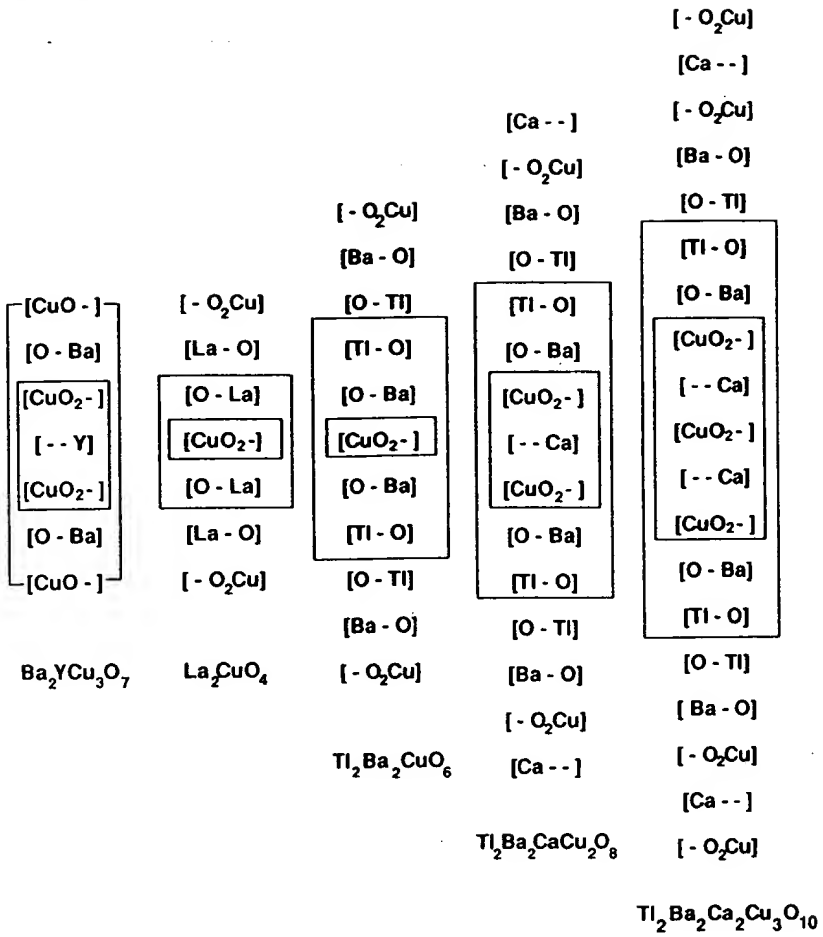


Figure 7.25 Layering schemes of various high-temperature superconductors. The CuO_2 plane layers are enclosed in small inner boxes, and the layers that make up a formula unit are enclosed in larger boxes. The Bi–Sr compounds $Bi_2Sr_2Ca_nCu_{n+1}O_{6+2n}$ have the same layering schemes as their Tl–Ba counterparts shown in this figure.

(i.e., body centered) planes repeat along the *c*-axis. It is these copper-oxide layers that are responsible for the superconducting properties.

A close examination of this figure shows that the general stacking rules mentioned in Section VI.C for the layering scheme are satisfied, namely metal ions in adjacent layers alternate between edge (E) and centered (C) sites, and adjacent layers never have oxygens on the same types of sites. The horizontal reflection symmetry at the central point of the cell is evident. It is also clear that $YBa_2Cu_3O_7$ is aligned and that the other four compounds are staggered.

Figure 7.26 (Torardi *et al.*, 1988a) presents a more graphical representation of the information in Fig. 7.25 by showing the positions of the atoms in their layers. The symmetry and body centering rules are also evident on this figure. Rao (1991) provided sketches for the six compounds $Tl_mBa_2Ca_nCu_{n+1}O_x$ similar to those in Fig. 7.26 with the compound containing one ($m = 1$) or two thallium layers ($m = 2$), where $n = 0, 1, 2$, as in the Torardi *et al.* figure.

B. Nomenclature

There are always two thalliums and two bariums in the basic formula for $Tl_2Ba_2Ca_nCu_{n+1}O_{6+2n}$, together with n calciums and $n + 1$ coppers. The first three members of this series for $n = 0$, 1, and 2 are called the 2201, 2212, and 2223 compounds, respectively, and similarly for their

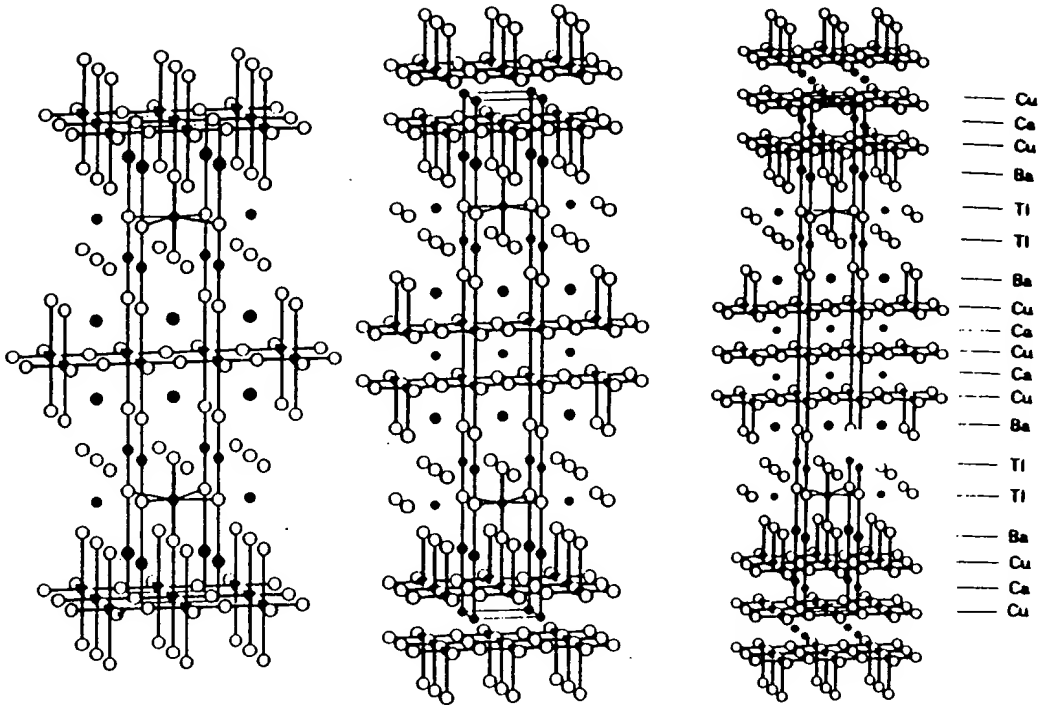


Figure 7.26 Crystal structures of $Tl_2Ba_2Ca_nCu_{n+1}O_{6+2n}$ superconducting compounds with $n = 0, 1, 2$ arranged to display the layering schemes. The $Bi_2Sr_2Ca_nCu_{n+1}O_{6+2n}$ compounds have the same respective structures (Torardi *et al.*, 1988a).

BiSr analogues $Bi_2Sr_2Ca_nCu_{n+1}O_{6+2n}$. Since Y in $YBa_2Cu_3O_7$ is structurally analogous to Ca in the Tl and Bi compounds, it would be more consistent to write $Ba_2YCu_3O_7$ for its formula, as noted in Section VI.F. In this spirit $Ba_2YCu_3O_{7-\delta}$ might be called the 0213 compound, and $(La_{1-x}M_x)_2CuO_{4-\delta}$ could be called 2001.

C. Bi–Sr Compounds

Now that the overall structures and interrelationships of the BiSr and TlBa high-temperature superconductors have been made clear in Figs. 7.25 and 7.26 we will comment briefly about each compound. Table 7.3 summarizes the characteristics of these and related compounds.

The first member of the BiSr series, the 2201 compound with $n = 0$, has octahedrally coordinated Cu and $T_c \approx 9$ K (Torardi *et al.*, 1988b). The second member, $Bi_2(Sr,Ca)_3Cu_2O_{8+\delta}$, is a superconductor with $T_c \approx 90$ K (Subramanian *et al.*, 1988a; Tarascon *et al.*, 1988b). There are two [Cu O_2 −] layers separated from each other by the [− − Ca] layer. The spacing from [Cu O_2 −] to [− − Ca] is 1.66 Å, which is less than the corresponding spacing of 1.99 Å between the levels [Cu O_2 −] and [− − Y] of YBaCuO. In both cases the copper ions have a pyramidal oxygen coordination of the type shown in Fig. 7.11. Superlattice structures have been reported along *a* and *b*, which means that minor modifications of the unit cells repeat approximately every five lattice spacings, as explained in Sect. IX.E. The third member of the series, $Bi_2Sr_2Ca_2Cu_3O_{10}$, has three CuO_2 layers separated from each other by [− − Ca] planes and a higher transition temperature, 110 K, when doped with Pb. The two Cu ions have pyramidal coordination, while the third is square planar.

Charge-density plots of

$$Bi_2Sr_2CaCu_2O_8$$

indicate the same type of covalency in the Cu–O bonding as with the $YBa_2Cu_3O_7$ and La_2CuO_4 compounds. They also indicate very little bonding between the adjacent [Bi – O] and [O – Bi] layers.

D. Tl–Ba Compounds

The TlBa compounds

$$Tl_2Ba_2Ca_nCu_{n+1}O_{6+2n}$$

have higher transition temperatures than their bismuth counterparts (Iqbal *et al.*, 1989; Subramanian *et al.*, 1988b; Torardi *et al.*, 1988a). The first member of the series, namely $Tl_2Ba_2CuO_6$ with $n = 0$, has no [– – Ca] layer and a relatively low transition temperature of ≈ 85 K. The second member ($n = 1$), $Tl_2Ba_2CaCu_2O_8$, called the 2212 compound, with $T_c = 110$ K has the same layering scheme as its Bi counterpart, detailed in Figs. 7.25 and 7.26. The [Cu O_2 –] layers are thicker and closer together than the corresponding layers of the bismuth compound (Toby *et al.*, 1990). The third member of the series, $Tl_2Ba_2Ca_2Cu_3O_{10}$, has three [Cu O_2 –] layers separated from each other by [– – Ca] planes, and the highest transition temperature, 125 K, of this series of thallium compounds. It has the same copper coordination as its BiSr counterpart. The 2212 and 2223 compounds are tetragonal and belong to the same crystallographic space group as La_2CuO_4.

We see from the charge-density plot of $Tl_2Ba_2CuO_6$ shown in Fig. 7.27 that Ba^{2+} is ionic, Cu exhibits strong covalency, especially in the Cu–O plane, and Tl also appears to have a pronounced covalency. The bonding between the [Tl – O] and [O – Tl] planes is stronger than that between the [Bi – O] and [O – Bi] planes of Bi–Sr.

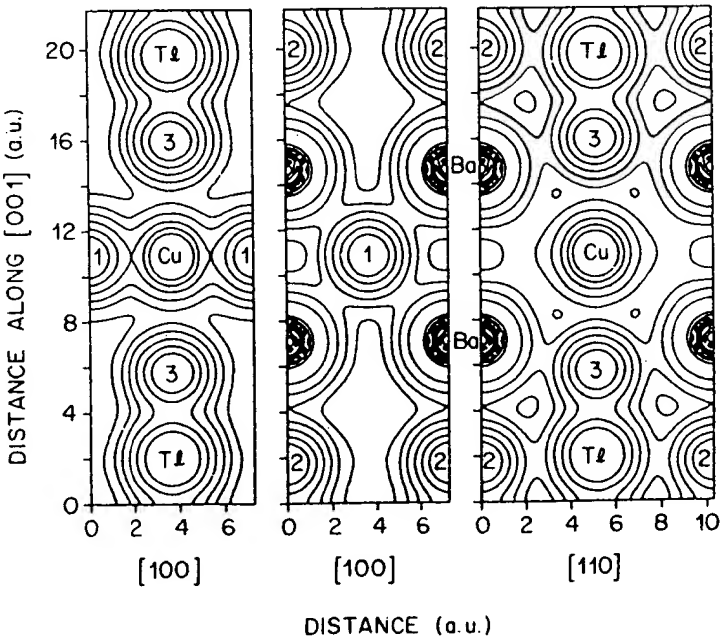


Figure 7.27 Contours of constant charge density on a logarithmic scale in two high-symmetry crystallographic planes of $Tl_2Ba_2CuO_6$. Oxygen atoms O(1), O(2), and O(3) are denoted 1, 2, and 3, respectively. The planar Cu–O1 binding is strongest (Hamann and Mattheiss, 1988; see Pickett, 1989).

E. Modulated Structures

The x-ray and neutron-diffraction patterns obtained during crystal structure determinations of the bismuth cuprates $Bi_2Sr_2Ca_nCu_{n+1}O_{6+2n}$ exhibit weak satellite lines with spacings that do not arise from an integral multiple of the unit cell dimensions. These satellites have modulation periods of 21 Å, 19.6 Å, and 20.8 Å, respectively, for the $n = 0$, 1, and 2 compounds (Li *et al.*, 1989). Since the lattice constant $a = 5.41$ Å ($b = 5.43$ Å) for all three compounds, this corresponds to a superlattice with unit cell of dimensions $\approx 3.8a$, b, c, with the repeat unit along the a direction equal to $\approx 3.8a$ for all three compounds. A modulation of $4.7b$ has also been reported (Kulik *et al.*, 1990). This structural modulation is called incommensurate because the repeat unit is not an integral multiple of a.

Substitutions dramatically change this modulation. The compound

$$Bi_2Sr_2Ca_{1-x}Y_xCu_2O_y$$

has a period that decreases from about $4.8b$ for $x = 0$ to the commensurate value $4.0b$ for $x = 1$ (Inoue *et al.*, 1989; Tamegai *et al.*, 1989). Replacing Cu by a transition metal (Fe, Mn, or Co) produces nonsuperconducting compounds with a structural modulation that is commensurate with the lattice spacing (Tarascon *et al.*, 1989b). A modulation-free bismuth–lead cuprate superconductor has been prepared (Manivannan *et al.*, 1991). Kistenmacher (1989) examined substitution-induced superstructures in $YBa_2(Cu_{1-x}M_x)_3O_7$. Superlattices with modulation wavelengths as short as 24 Å have been prepared by employing ultra-thin deposition techniques to interpose insulating planes of $PrBa_2Cu_3O_7$ between superconducting Cu–O layers of $YBa_2Cu_3O_7$ (Jakob *et al.*, 1991; Lowndes *et al.*, 1990; Pennycook *et al.*, 1991; Rajagopal and Mahanti, 1991; Triscone *et al.* 1990). Tanaka and Tsukada (1991) used the Kronig–Penney model (Tanaka and Tsukada, 1989a, b) to calculate the quasiparticle spectrum of superlattices.

F. Aligned Tl–Ba Compounds

A series of aligned thallium-based superconducting compounds that have the general formula $TlBa_2Ca_nCu_{n+1}O_{5+2n}$ with n varying from 0 to 5 has been reported (Ihara *et al.*, 1988; Rona, 1990). These constitute a series from 1201 to 1245. They have superconducting transition temperatures almost as high as the $Tl_2Ba_2Ca_nCu_{n+1}O_{6+2n}$ compounds. Data on these compounds are listed in Table 7.3.

G. Lead Doping

In recent years a great deal of effort has been expended in synthesizing lead-doped superconducting cuprate structures (Itoh and Uchikawa, 1989). Examples involve substituting Pb for Bi (Dou *et al.*, 1989; Zhengping *et al.*, 1990), for Tl (Barry *et al.*, 1989; Mingzhu *et al.*, 1990), or for both Bi and Tl (Iqbal *et al.*, 1990). Different kinds of Pb, Y-containing superconductors have also been prepared (cf. Mattheiss and Hamann, 1989; Ohta and Maekawa, 1990; Tang *et al.*, 1991; Tokiwa *et al.*, 1990, 1991).

X. ALIGNED HgBaCaCuO

The series of compounds

$$HgBa_2Ca_nCu_{n+1}O_{2n+4},$$

where n is an integer, are prototypes for the Hg family of superconductors. The first three members of the family, with $n = 0, 1, 2$, are often referred to as Hg-1201, Hg-1212, and Hg-1223, respectively. They have the structures sketched in Fig. 7.28 (Tokiwa-Yamamoto *et al.*, 1993; see also Martin *et al.*, 1994; Putilin *et al.*, 1991). The lattice constants are $a = 3.86$ Å for all of them, and $c = 9.5$, 12.6, and 15.7 Å for $n = 0, 1, 2$, respectively. The atom positions of the $n = 1$ compound are listed in Table 7.6 (Hur *et al.*, 1994). The figure is drawn

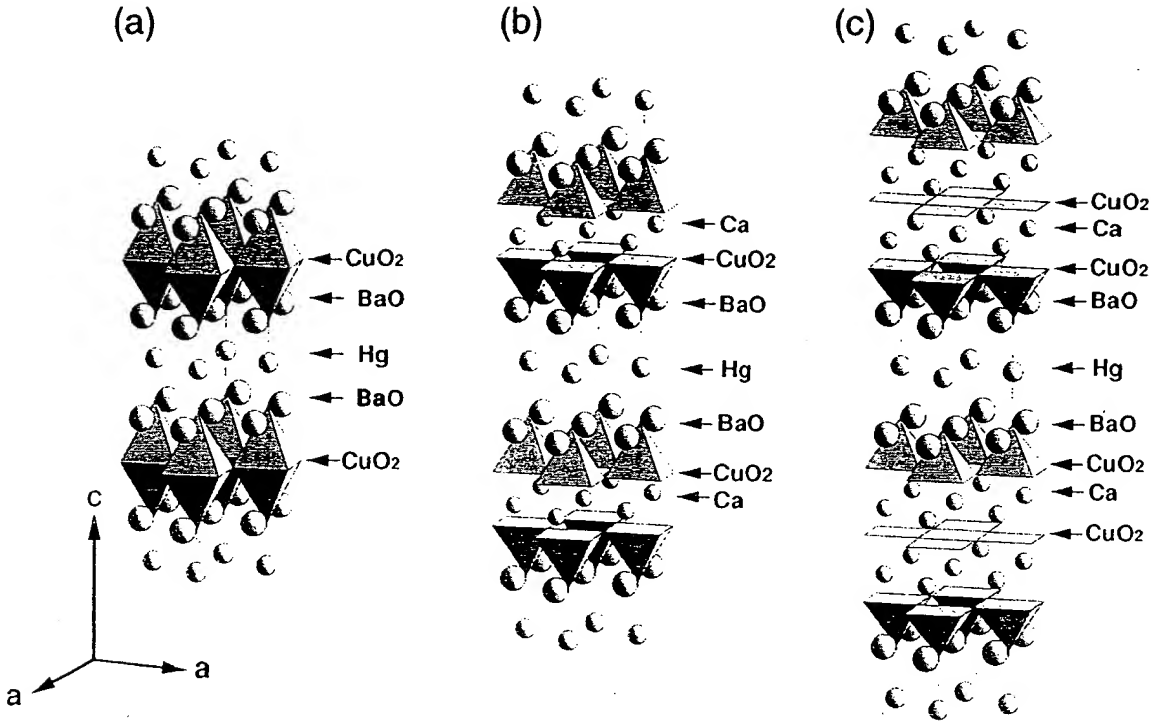


Figure 7.28 Structural models for the series $HgBa_2Ca_nCu_{n+1}O_{2n+4}$. The first three members with $n = 0, 1, 2$ are shown (parts a, b, and c, respectively) (Tokiwa-Yamamoto *et al.*, 1993).

Table 7.6 Normalized Atom Positions in the Tetragonal Unit Cell of $HgBa_2Ca_{0.86}Sr_{0.14}Cu_2O_{6+\delta}$ [a]

Layer	Atom	x	y	z
	Hg	0	0	1
[Hg – –]				
	O(3)	$\frac{1}{2}$	$\frac{1}{2}$	1
	O(2)	0	0	0.843
[O – Ba]				
	Ba	$\frac{1}{2}$	$\frac{1}{2}$	0.778
	Cu	0	0	0.621
[Cu O_2 –]	O(1)	0	$\frac{1}{2}$	0.627
	O(1)	$\frac{1}{2}$	0	0.627
[– – Ca]	Ca, Sr	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$
	O(1)	$\frac{1}{2}$	0	0.373
[Cu O_2 –]	O(1)	0	$\frac{1}{2}$	0.373
	Cu	0	0	0.379
	Ba	$\frac{1}{2}$	$\frac{1}{2}$	0.222
[O – Ba]				
	O(2)	0	0	0.157
	O(3)	$\frac{1}{2}$	$\frac{1}{2}$	0
[Hg – –]				
	Hg	0	0	0

[a] Unit cell dimensions $a = 3.8584$ Å and $c = 12.6646$ Å, space group is $P4/mmm$, D_{4h}^1. The Hg site is 91% occupied and the O(3) site is 11% occupied ($\delta = 0.11$). The data are from Hur *et al.* (1994).

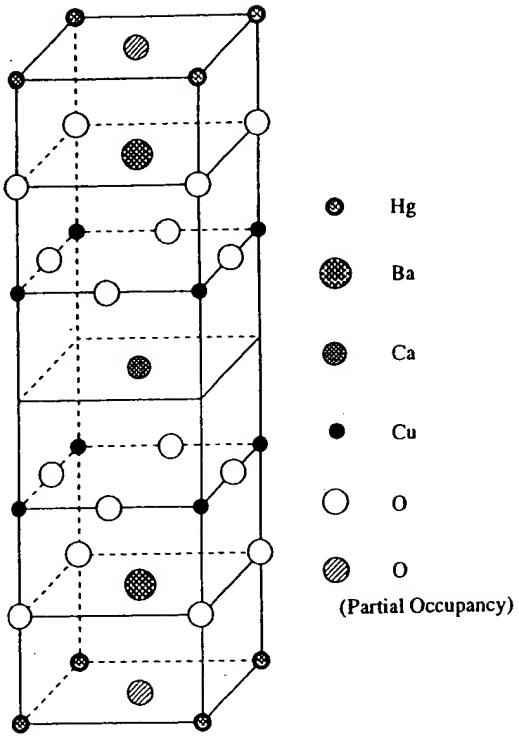


Figure 7.29 Schematic structure of the $HgBa_2CaCu_2O_{6+\delta}$ compound which is also called Hg-1212 (Meng *et al.*, 1993a).

with mercury located in the middle layer of the unit cell, while the table puts Hg at the origin (000) and Ca in the middle ($\frac{1}{2}\frac{1}{2}\frac{1}{2}$). Figure 7.29 presents the unit cell for the $n = 1$ compound $HgBa_2CaCu_2O_{6+\delta}$ drawn with Ca in the middle (Meng *et al.*, 1993a). The symbol δ represents a small excess of oxygen located in the center of the top and bottom layers, at positions $\frac{1}{2}\frac{1}{2}0$ and $\frac{1}{2}\frac{1}{2}1$ which are labeled "partial occupancy" in the figure. If this oxygen were included the level symbol would be [Hg – O] instead of [Hg – –]. These Hg compound structures are similar to those of the series $TlBa_2Ca_nCu_{n+1}O_{2n+4}$ mentioned above in Section IX.F.

We see from Fig. 7.28 that the copper atom of Hg-1201 is in the center of a stretched octahedron with the planar oxygens O(1) at a distance of 1.94 Å, and the apical oxygens O(2) of the [O – Ba] layer much further away (2.78 Å). For $n = 1$ each copper atom is in the center of the

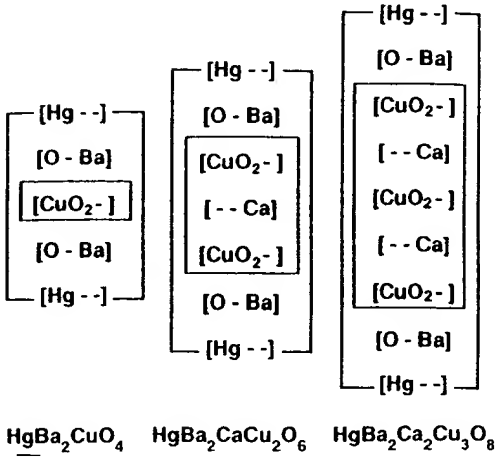


Figure 7.30 Layering schemes of three $HgBa_2Ca_nCu_{n+1}O_{2n+4}$ compounds, using the notation of Fig. 7.25.

base of a tetragonal pyramid, and for $n = 2$ the additional CuO_2 layer has Cu atoms which are square planar coordinated. The layering scheme stacking rules of Section VI.C are obeyed by the Hg series of compounds, with metal ions in adjacent layers alternating between edge (E) and centered (C) sites, and oxygen in adjacent layers always at different sites. We see from Table 7.6 that the [O – Ba] layer is strongly puckered and the [Cu O_2 –] layer is only slightly puckered.

The relationships between the layering scheme of the $HgBa_2Ca_nCu_{n+1}O_{2n+4}$ series of compounds and those of the other cuprates may be seen by comparing the sketch of Fig. 7.30 with that of Fig. 7.25. We see that the $n = 1$ compound $HgBa_2CaCu_2O_6$ is quite similar in structure to $YBa_2Cu_3O_7$ with Ca replacing Y in the center and Hg replacing the chains [Cu O –]. More surprising is the similarity between the arrangement of the atoms in the unit cell of each

$$HgBa_2Ca_nCu_{n+1}O_{2n+4}$$

compound and the arrangement of the atoms in the semi-unit cell of the corresponding

$$Tl_2Ba_2Ca_nCu_{n+1}O_{2n+6}$$

compound. They are the same except for the replacement of the [Tl – O] layer by [Hg – –], and the fact that the thallium compounds are body centered and the Hg ones are aligned.

Supercells involving polytypes with ordered stacking sequences of different phases, such as Hg-1212 and Hg-1223, along the c direction have been reported. The stoichiometry is often

$$Hg_2Ba_4Ca_3Cu_5O_x$$

corresponding to equal numbers of the Hg-1212 and Hg-1223 phases (Phillips, 1993; Schilling *et al.*, 1993, 1994).

Detailed structural data have already been reported on various Hg family compounds such as $HgBa_2CuO_{4+\delta}$ (Putlin *et al.*, 1993) and the $n = 1$ compound with partial Eu substitution for Ca (Putlin *et al.*, 1991). The compound

$$Pb_{0.7}Hg_{0.3}Sr_2Nd_{0.3}Ca_{0.7}Cu_3O_7$$

has Hg in the position (0.065, 0, 0), slightly displaced from the origin of the unit cell (Martin *et al.*, 1994). Several researchers have reported synthesis and pretreatment procedures (Adachi *et al.*, 1993; Itoh *et al.*, 1993; Isawa 1994a; Meng, 1993b; Paranthaman, 1994; Paranthaman *et al.*, 1993). Lead doping for Hg has been used to improve the superconducting properties (Iqbal *et al.*, 1994; Isawa *et al.*, 1993; Martin *et al.*, 1994).

XI. BUCKMINSTERFULLERENES

The compound C_{60}, called buckminsterfullerene, or fullerene for short, consists of 60 carbon atoms at the vertices of the dotriacontohedron (32-sided figure) that is sketched in Fig. 3.35 and discussed in Chapter 3, Section XVI. The term fullerene is used here for a wider class of compounds C_n with n carbon atoms, each of whose carbon atoms is bonded to three other carbons to form a closed surface, with the system conjugated such that for every resonant structure each carbon has two single bonds and one double bond. The smallest possible compound of this type is tetrahedral C_4, which has the three resonant structures shown in Fig. 7.31. Cubic C_8 is a fullerene, and we show in Problem 17 that it has nine resonant structures. Icosahedral C_{12} is also a fullerene, but octahedral C_6 and dodecahedral C_{20} are not because their carbons are bonded to more than three neighbors. These hypothetical smaller C_n compounds have never been synthesized, but the larger ones, such as C_{60}, C_{70}, C_{76}, C_{78}, and C_{82}, have been made and characterized. Some of them have several forms, with different arrangements of polygons. Clusters of buckminsterfullerenes, such as icosahedral $(C_{60})_{13}$, have also been studied (T. P. Martin *et al.*, 1993).

There are several interesting geometrical characteristics of fullerenes (Chung and Sternberg, 1993). Since each carbon (vertex) joins three bonds (edges) and each edge has two vertices, the number of edges E in a structure C_n is 50% greater than the number of vertices V. There is a general theorem in topology, called Euler's Theorem, that the number of faces F of a

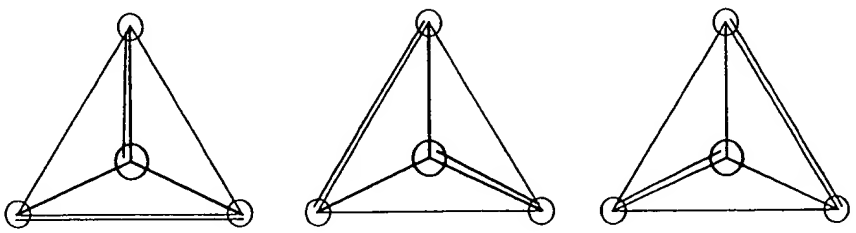


Figure 7.31 The three resonant structures of the (hypothetical) tetrahedral compound C_4.

polyhedron is given by the formula

$$F = E - V + 2. \tag{7.8}$$

In a fullerene C_n where $n = V$ three edges meet at each vertex, so we have

$$E = 3V/2, \tag{7.9}$$

$$F = \frac{V}{2} + 2. \tag{7.10}$$

It is shown in Problem 16 that

$$E = \tfrac{1}{2}\sum_s sF_s \tag{7.11a}$$

$$V = \tfrac{1}{3}\sum_s sF_s, \tag{7.11b}$$

where F_s is the number of faces with s sides, and of course,

$$F = \sum_s F_s. \tag{7.12}$$

Combining Eqs. (7.10)–(7.12) gives the fullerene face formula

$$\sum_s (6 - s)F_s = 12. \tag{7.13}$$

This expression does not place any restrictions on the number of hexagons (F_s), but it does severely limit the number of other polyhedra. The two smallest hypothetical fullerenes, the tetrahedron and the cube, have no hexagons, and the larger ones consist of 12 pentagons (F_s), from Eq. (7.13), and numerous hexagons. For example, the molecule C_{60} with $V = 60$ has 12 pentagons and 20 hexagons. Table 7.7 gives the geometric characteristics of the five Platonic solids, the solids generated by truncating all of their vertices, and several other regular polygons, most of which are fullerenes. The fullerenes of current interest are C_{60} and larger molecules consisting of 12 pentagons and numerous hexagons, such as C_{70}, C_{76}, C_{78}, and C_{82}. Some have several varieties, such as the isomers of C_{78} with the symmetries C_{2v}, D_3, and D_{3h} (Diederich and Whetten, 1992).

The outer diameter of the C_{60} molecule is 7.10 Å and its van der Waals separation is 2.9 Å, so that the nearest-neighbor distance (effective diameter) in a

Table 7.7 Characteristics of Several Regular Solids[a]

Figure	Vertices	Edges	Faces	Face (polygon) type
Tetrahedron	4	6	4	all equilateral triangles
Octahedron[b]	6	12	8	all equilateral triangles
Cube	8	12	6	all squares
Icosahedron[b]	12	30	20	all equilateral triangles
Dodecahedron (pentagonal)	20	30	12	all regular pentagons
Hexadecahedron	28	42	16	12 pentagons, 4 hexagons
Truncated tetrahedron	12	18	8	4 equilateral triangles, 4 hexagons
Truncated octahedron	24	36	14	6 squares, 8 hexagons
Truncated cube	24	36	14	8 equilateral triangles, 6 octagons
Dotriacontohedron (truncated icosahedron)	60	90	32	12 regular pentagons, 20 hexagons
Truncated dodecahedron	60	90	32	20 equilateral triangles, 12 decagons
Heptatriacontohedron	70	105	37	12 pentagons (2 regular), 25 hexagons
Tetracontahedron	76	114	40	12 pentagons, 28 hexagons
Hentetracontohedron	78	116	41	12 pentagons, 29 hexagons
Dotetracontohedron	84	126	44	12 pentagons, 32 hexagons
Large Fullerene	n	$\frac{3}{2}n$	$\frac{1}{2}n + 2$	12 pentagons, $\frac{1}{2}n - 10$ hexagons

[a] The first five solids are the Platonic solids, and the seventh to eleventh are truncations of the Platonic solids. When carbons occupy the vertices all correspond to fullerenes except the octahedron and the icosahedron for which $3V \neq 2E$. The smallest compounds in this table have never been synthesized.

[b] Not a fullerene because the vertices have more than three edges.

solid is 10.0 Å. The bonds shared by a five-membered and a six-membered ring are 1.45 Å long, while those between two adjacent six-membered rings are 1.40 Å long. Above 260 K these molecules form a face centered cubic lattice with lattice constant 14.2 Å; below 260 K it is simple cubic with $a = 7.10$ Å (Fischer *et al.*, 1991; Kasatani *et al.*, 1993; Troullier and Martins, 1992). When C_{60} is doped with alkali metals to form a superconductor it crystallizes into a face centered cubic lattice with larger octahedral and smaller tetrahedral holes for the alkalis. The C_{60} ions are orientationally disordered in the lattice (Gupta and Gupta, 1993).

XII. SYMMETRIES

Earlier in this chapter we mentioned the significance of the horizontal reflection plane σ_h characteristic of the high-temperature superconductors, and noted that most of these superconductors are body centered. In this section we will point out additional symmetries that are present. Table VI-14 of our earlier work (Poole *et al.*, 1988) lists the point symmetries at the sites of the atoms in a number of these compounds.

In the notation of group theory the tetragonal structure belongs to the point group $4/mmm$ (this is the newer international notation for what in the older Schönflies notation was written D_{4h}). The unit cell possesses the inversion operation at the center, so when there is an atom at position (x, y, x), there will be another identical atom at position $(-x, -y, -z)$. The international symbol $4/mmm$ indicates the presence of a fourfold axis of symmetry C_4 and three mutually perpendicular mirror planes m. The Schönflies notation D_{4h} also specifies the fourfold axis, h signifying a horizontal mirror plane σ_h and D indicating a dihedral group with vertical mirror planes.

We see from Fig. 7.32 that the z-axis is a fourfold (90°) symmetry axis called C_4, and that perpendicular to it are twofold (180°) symmetry axes along the x and y directions, called C_2, and also along the diagonal directions (C_2') in the midplane. There are two vertical mirror planes σ_v, two diagonal mirror planes σ_d which are also vertical, and a horizontal mirror plane σ_h. Additional symmetry operations that are not shown are a 180° rotation C_2^z around the z axis,

$$C_2^z = C_4^z C_4^z, \tag{7.14}$$

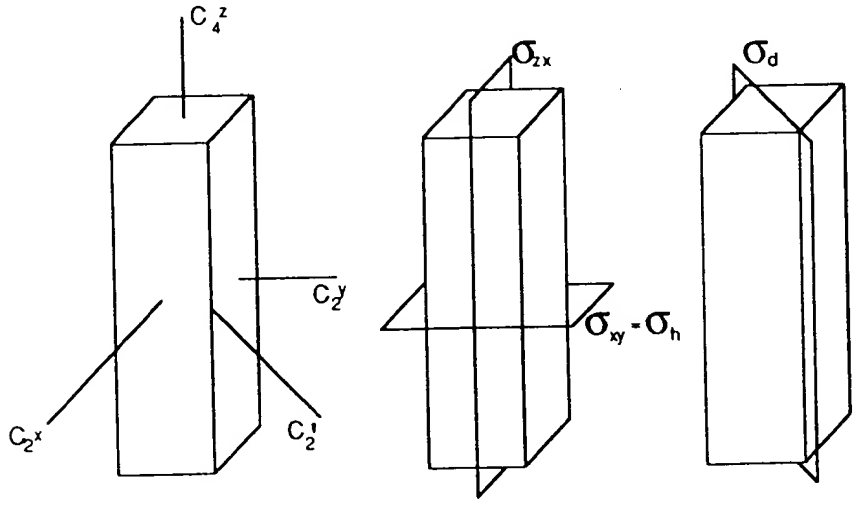


Figure 7.32 Symmetry operations of the tetragonal unit cell showing a fourfold rotation axis C_4, three twofold axes C_2, and reflection planes of the vertical $\sigma_{zx} = \sigma_v$, horizontal $\sigma_{xy} = \sigma_h$; and diagonal σ_d types.

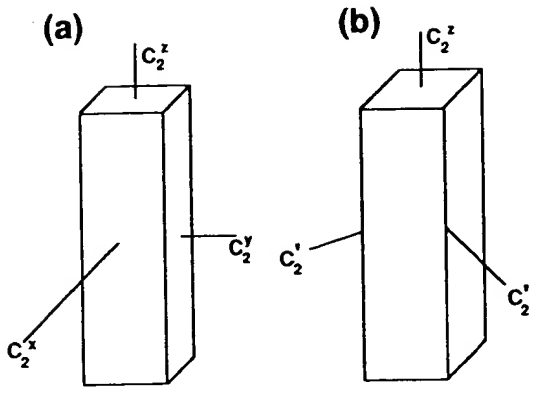


Figure 7.33 Rotational symmetry operations of an orthorhombic unit cell (a) with rectangular distortion, and (b) with rhombal distortion from an originally tetragonal cell.

and the improper fourfold rotation S_4^z around z that corresponds to C_4^z followed by, or preceded by, σ_h,

$$S_4^z = C_4^z \sigma_h = \sigma_h C_4^z, \qquad (7.15)$$

where C_4^z and σ_h commute.

The orthorhombic structure has *mmm*, D_{2h} symmetry. We see from Fig. 7.33 that both the rectangular and rhombal unit cells, which correspond to Figs. 7.4a and 7.4b, respectively, have three mutually perpendicular twofold axes, and that they also have three mutually perpendicular mirror planes σ, which are not shown. The two cases differ in having their horizontal axes and vertical planes oriented at 45° to each other.

Cubic structures, being much higher in symmetry, have additional symmetry operations, such as fourfold axes C_4^x, C_4^y, and C_4^z along each coordinate direction, threefold axes C_3 alorig each body diagonal, and numerous other mirror planes. These can be easily seen from an examination of Fig. 7.1. Buckyballs belong to the icosohedral group, which has twofold (C_2), fivefold (C_5), and sixfold (C_6) rotation axes, horizontal reflection planes, inversion symmetry, and sixfold (S_6) and tenfold (S_{10}) improper rotations, for a total of 120 individual symmetry operations in all (Cotton, 1963).

XIII. CRYSTAL CHEMISTRY

In Chapter 3 we briefly described the structures of some classical superconductors, and in this chapter we provided a more detailed discussion of the structures of the cuprate superconductors. The question arises of how structure is related to the presence of metallic and superconducting properties.

Villars and Phillips (1988; Phillips, 1989a) proposed to explain the combinations of elements in compounds that are favorable for superconductivity at relatively high temperatures by assigning three metallic coordinates to each atom, namely an electron number N_e, a size r, and an electronegativity X. The electron numbers are given in Table 3.1 for most of the elements, with $N_e = 3$ for all of the rare earths and actinides; several correlations of N_e with T_c have already been given in Chapter 3. The sizes and electronegativities were determined empirically from a study of some 3,000 binary intermetallic compounds of types AB, AB_2, AB_3, and A_2B_5. The resulting values for each atom are listed in Fig. 7.34 together with their electron numbers. These values, although arrived at empirically on the basis of the constraint of self-consistency, do have a spectroscopic basis, and thus are called, respectively, *spectroscopic radii* and *spectroscopic electronegativities*.

The metallic coordinates of the atoms can be employed to calculate the three Villars–Phillips (VP) coordinates for each compound, namely (a) average number of valence electrons $N_v = \langle N_e \rangle_{av}$, (b) spectroscopic electronegativity difference ΔX, and (c) spectroscopic radius difference ΔR, where we are using the VP notation. For example, for the compound NbN, with $T_c = 17.3$ K, we have, using the data from Fig. 7.34,

$$\begin{aligned} N_v &= \tfrac{1}{2}(4+5) = 4.5, \\ \Delta R &= 2.76 - 0.54 = 2.22, \\ \Delta X &= 2.03 - 2.85 = -0.82. \end{aligned} \qquad (7.16)$$

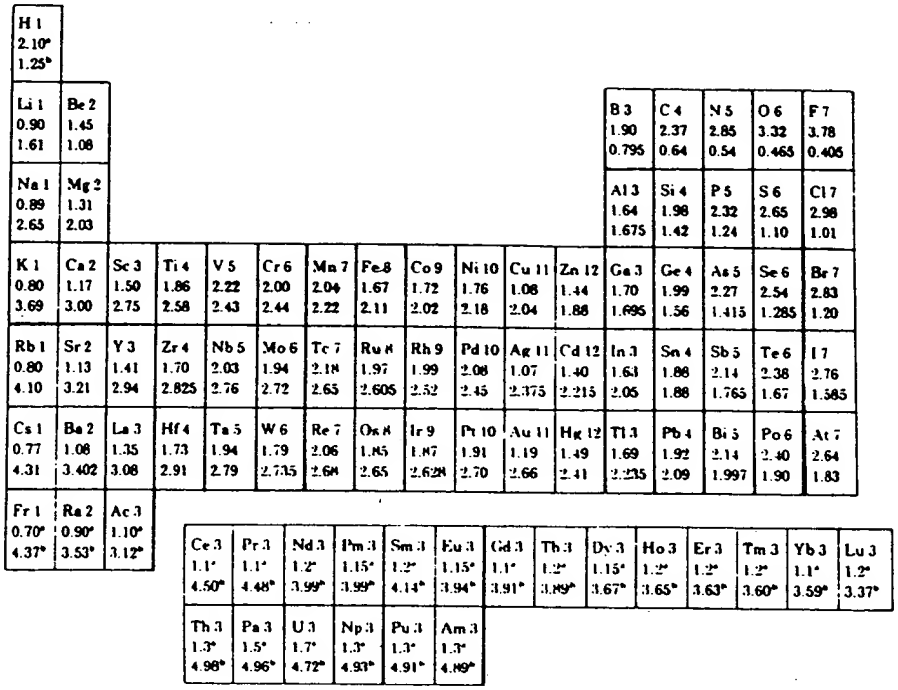


Figure 7.34 Periodic table listing metallic valences (upper right), sizes (center), and electronegativities (bottom) in the box of each element, according to the Villars–Phillips model (Phillips, 1989a, p. 321).

The VP coordinates for the $A15$ compound Ge_3Nb with $T_c = 23.2$ K are calculated as follows:

$$N_v = \tfrac{1}{4}(4 + 3 \times 5) = 4.75,$$

$$\Delta R = \tfrac{1}{2}(1.56 - 2.76) = -0.60, \quad (7.17)$$

$$\Delta X = \tfrac{1}{2}(1.99 - 2.03) = -0.02.$$

The text by Phillips (1989a) tabulates the VP coordinates for more than 60 superconductors with $T_c > 10$ K and for about 600 additional superconductors with transition temperatures in the range $1 < T_c < 10$ K.

When the points for the 600 compounds with lower transition temperatures are plotted on a three-dimensional coordinate system with axes N_v, ΔX, and ΔR, they scatter over a large range of values, but when the points for compounds with $T_c > 10$ K are plotted, they are found to cluster in three regions, called islands, as shown in Fig. 7.35. Island A contains the $A15$ compounds plus some complex intermetallics, island B consists mainly of the NbN family plus some borides and carbides, and island C has closely clustered Chevrel phases, with the high-T_c cuprates on the left. When ternary ferroelectric oxides with Curie temperatures that exceed 500°C are plotted in the same diagram as the superconductors they cluster between the Chevrel group and the cuprates. These ferroelectric oxides are not superconductors, though Phillips (1989a) suggested that doping them with Cu and alkaline earths could produce superconductors with high transition temperatures.

Thus we see that the high transition temperatures of classical superconductors are favored by particular structures and by particular combinations of metallic coordinates for each of these structures. The Villars–Phillips approach provides both structural and atomic criteria for the presence of high T_c.

We have discussed the Phillips approach to a crystal chemistry explanation of the superconductivity of the cuprates. Other researchers have offered alternate, in some cases somewhat related, approaches to understanding the commonali-

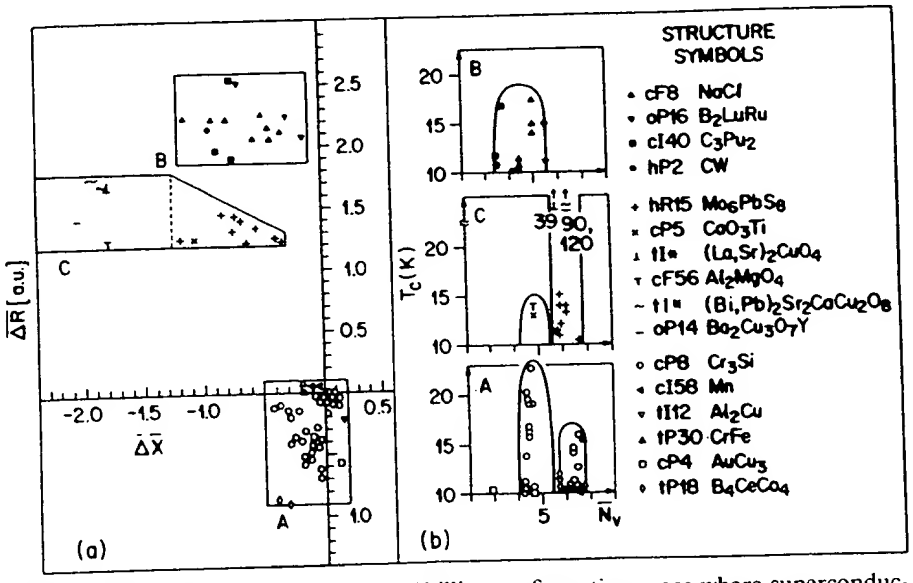


Figure 7.35 Regions in the Villars–Phillips configuration space where superconductivity occurs at relatively high temperatures (Phillips, 1989a, p. 324; Villars and Phillips, 1988).

ties of the various high-temperature and classical superconductors (Adrian, 1992; Schneider, 1992; Tajima and Kitazawa, 1990; Whangbo and Torardi, 1991; Torrace, 1992; Yakhmi and Iyer, 1992; Zhang and Sato, 1993).

XIV. COMPARISON WITH CLASSICAL SUPERCONDUCTOR STRUCTURES

Many elements such as copper and lead are face centered cubic, while many other elements, such as niobium, are body centered cubic, with $a = 3.30$ Å for Nb. The $A15$ compounds, such as Nb_3Se, are (simple) cubic with lattice constant $a \approx 3.63\sqrt{2}$ and have parallel chains of Nb atoms 5.14 Å apart. Other types of classical superconductors, such as the Laves and Chevrel phases, are cubic or close to cubic. The new oxide superconductors are tetragonal or orthorhombic close to tetragonal, and they all have $a \approx b \approx 3.85$ Å, which is somewhat greater than the value for the $A15$ compounds. The third lattice constant c varies with the compound, with the values 13.2 Å for LaSrCuO, 11.7 Å for YBaCuO, and ≈ 23 to 36 Å for the BiSrCaCuO and TlBaCaCuO compounds. These differences occur because the number of copper–oxygen and other planes per unit cell, as well as the spacings between them, vary from compound to compound due to the diverse arrangements of atoms between the layers. Thus relatively high-symmetry crystal structures are characteristic of many superconductors.

XV. CONCLUSIONS

Almost all the high-temperature oxide superconductors have point symmetry D_{4h} ($a = b$) or symmetry close to D_{4h} ($a \approx b$). These superconductors consist of horizontal layers, each of which contains one positive ion and either zero, one, or two oxygens. The copper ions may be coordinated square planar, pyramidal, or octahedral, with some additional distortion. Copper oxide layers are never adjacent to each other, and equivalent layers are never adjacent. The cations alternate sites vertically, as do the oxygens. The copper oxide layers are either flat or slightly puckered, in contrast to the other metal oxide layers, which are generally far from planar. The highest T_c compounds have metal layers

(e.g., Ca) with no oxygens between the copper oxide planes.

FURTHER READING

The Wyckoff series, *Crystal Structures* (1963, Vol. 1; 1964, Vol. 2; 1965, Vol. 3; 1968, Vol. 4) provides a comprehensive tabulation of crystal structures, but many important classical superconductors such as the $A15$ compounds are not included. The *International Tables for X-Ray Crystallography* (Henry and Lonsdale, 1965, Vol. 1) provide the atom positions and symmetries for all of the crystallographic space groups. The Strukturbericht notation, e.g., $A15$ for Nb_3Ge, is explained in Pearson's compilation (1958).

Details of cuprate crystallographic structures are given by Beyers and Shaw (1989; $YBa_2Cu_3O_7$), Burns and Glazer (1990), Hazen (1990), Poole *et al.* (1988, Chapter 6), Santoro (1990), and Yvon and François (1989). Phillips (1989a) provides an extensive discussion of the crystal chemistry of the cuprates. Our earlier work (Poole *et al.*, 1988, p. 107) lists the site symmetries in perovskite and cuprate structures. Billinge *et al.* (1994) reviewed lattice effects in high temperature superconductors, and Zhu (1994) reviewed structural defects in $YBa_2Cu_3O_{7-\delta}$.

The microstructure of high temperature superconductors studied by electron microscopy are reviewed by Chen (1990), Gai and Thomas (1992), Gross and Koelle (1994), and Shekhtman (1993). Oxygen stoichiometry in HTSC's is reviewed by Chandrashekhar *et al.*, (1994), Green and Bagley (1990) and by Routbert and Rothman (1995). Electron-doped superconductors are reviewed by Almasan and Maple (1991) and by Fontcuberta and Fàbrega (1995).

The March 1992 special issue of *Accounts of Chemical Research* (Vol. 25, No. 3) is devoted to reviews of buckminsterfullerenes. Two recent books are edited by Billups and Ciofolini (1993) and by Kroto and Walton (1993), and the review by Dresselhaus *et al.* (1994) are devoted to fullerenes. The thallium compounds were reviewed by Hermann and Yakhimi (1993) and the mercury superconductors by Chu (1995).

PROBLEMS

1. Show that the radius of the octahedral hole in an fcc close-packed lattice of atoms of radius r_0 is equal to $[\sqrt{2} - 1]r_0$. What is the radius of the hole if the lattice is formed from oxygen ions?
2. Show that the radius of the tetrahedral hole in an fcc close-packed lattice of atoms of radius r_0 is equal to $[(3/2)^{1/2} - 1)r_0$. What is the radius of the hole if the lattice is formed from oxygen ions?
3. The "image perovskite" unit cell is generated from the unit cell of Fig. 7.1 by shifting the origin from the point $(0,0,0)$ to the point $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$. Sketch this "image" cell. Show that the planes of atoms in this cell are the image planes related by the body centering operation to those of the original perovskite. This image cell is the one that usually appears to represent perovskite in solid-state physics texts.
4. Calculate the distance between the yttrium atom and its nearest-neighbor Ba, Cu, and O atoms in the superconductor $YBa_2Cu_3O_7$.
5. Write down the x, y, z coordinates for the five numbered atoms in the initial plane of Fig. 7.16. Give the explicit symmetry operations, with the proper choice of sign in Eq. (7.5) for each case, that transform these five atoms to their indicated new positions on the other three planes.
6. Explain how the international and Schönflies symbols, *mmm* and D_{2h} respectively, are appropriate for designating the point group for the orthorhombic superconductors.
7. What are the symmetry operations of the $A15$ unit cell of Fig. 3.19?
8. The D_{2h} point group consists of eight symmetry operations that leave an orthorhombic cell unchanged, namely an identity operation E that produces no change, three twofold rotations C_2^i along $i = x, y, z$, three mirror reflection planes σ_{ij}, and an inversion i. Examples of these symmetry operations are

$$\begin{array}{llll} E & x \to x & y \to y & z \to z \\ C_2^x & x \to x & y \to -y & z \to -z \\ \sigma_{xy} & x \to x & y \to y & z \to -z \\ i & x \to -x & y \to -y & z \to -z. \end{array}$$

A group has the property that successive application of two symmetry oper-

ations produces a third. Thus, we have, for example,

$$C_2^x \sigma_{xy} = \sigma_{zx}$$
$$C_2^y C_2^x = C_2^z$$
$$iC_2^y = \sigma_{zx}$$
$$\sigma_{zx}\sigma_{yz} = C_2^z.$$

These results have been entered into the following multiplication table for the D_{2h} group. Fill in the remainder of the table. Hint: each element of a group appears in each row and each column of the multiplication table once and only once.

	E	C_2^x	C_2^y	C_2^z	i	σ_{xy}	σ_{yz}	σ_{zx}
E						σ_{zx}		
C_2^x								
C_2^y		C_2^z						
C_2^z								
i			σ_{zx}					
σ_{xy}								
σ_{yz}								
σ_{zx}							C_2^z	

9. Construct the multiplication table for the D_{4h} point group which contains the 16 symmetry elements that leave a tetragonal unit cell unchanged. Which pairs of symmetry elements A and B do not commute, i.e., such that $AB \neq BA$? Hint: follow the procedures used in Problem 8.
10. Draw diagrams analogous to those in Fig. 7.25 for the first two members of the aligned series $TlBa_2Ca_nCu_{n+1}O_{5+2n}$, where $n = 0, 1$.
11. Draw the analogue of Fig. 7.19 for the Nd_2CuO_4 compound, showing the location of all of the Cu and O atoms. How do Figs. 7.21 and 7.22 differ for Nd_2CuO_4?
12. Calculate the Villars–Phillips coordinates for the three superconductors MoP_3, V_3Sn, and NbTi.
13. Select one of the compounds ($Tl_2Ba_2CuO_6$, $Bi_2Sr_2CaCu_2O_8$, $Bi_2Sr_2Ca_2Cu_3O_{10}$, $Tl_2Ba_2Ca_2Cu_3O_6$) and construct a table for it patterned after Tables 7.5 or 7.6.
14. Locate a twofold (C_2), fivefold (C_5), and sixfold (C_6) rotation axis, and also a reflection plane σ_h in the buckyball sketch of Fig. 3.35. How many of each type of operation are there?
15. We can see by examining Fig. 3.35 that a buckyball has inversion symmetry. Identify a sixfold (S_6) and tenfold (S_{10}) improper rotation axis, where an improper rotation is understood to involve a sequential inversion and a proper rotation. How many S_6 and how many S_{10} axes are there?
16. Show that the total number of edges E in a fullerene is given by

$$E = \tfrac{1}{2}\sum_s sF_s,$$

and the number of vortices is

$$V = \tfrac{1}{3}\sum_s sF_s,$$

where F_s is the number of faces with s sides.
17. Show that the cubic fullerene compound C_8 has nine resonant structures.

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